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An Evaluation of the Role of Ozone, Acid Deposition, and other Airborne Pollutants in the Forests of Eastern North America

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An Evaluation of the Role of Ozone, Acid Deposition, and Other Airborne Pollutants in the Forests of Eastern North America

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INTRODUCTION

Purpose of this Document

In recent years, there has been increasing scientific and public concern that airborne pollutant chemicals might be causing some of the visible injuries and decreases in growth of forests that have been reported in various parts of North America. This concern was further stimulated by reports of widespread visible symptoms and increased mortality of forests in central Europe. Sulfur dioxide, ozone, nitrogen oxides and acid deposition are all suspected to be among the airborne pollutant chemicals that could be involved.

One of the major objectives of current forestry research is to determine if wet and dry deposition of acidic and acidifying substances derived from short- and long-distance transport of airborne sulfur and nitrogen oxides could account for the changes in the condition of forests observed in the eastern United States. These changes include decreased growth, increased visible symptoms of injury, and mortality of high-elevation spruce-fir forests; increased visible injury and mortality of sugar maple and other eastern hardwood forests; and decreased radial growth of southern pine forests without visible symptoms of injury. This objective has it origins in the public and scientific concern about "acid rain." Acid deposition is known to be causing detrimental effects in aquatic ecosystems; however, convincing scientific evidence that acid deposition is also harming forests is lacking.

The Forest Response Program (FRP) of the National Acid Precipitation Assessment Program (NAPAP) is a major interagency effort initiated specifically to investigate effects of acidic deposition and associated pollutants on forests and forest tree species. The Southern Commercial Forest Research Cooperative functions under this umbrella. This document was conceived to assist the Cooperative in resolving questions relating to the prevailing scientific judgement that ozone and other photochemical oxidants, either alone or in combination with acid deposition, are the airborne pollutant chemicals most likely to affect southern commercial forests, eastern hardwood forests, and possibly also eastern spruce-fir forests; and the prevailing public perception that emissions of sulfur and nitrogen oxides leading to increased deposition of acidic and acidifying substances are the airborne pollutant chemicals most likely to adversely affect these same forests.

Approach

The approach used in preparing this document has been to develop a critical review of published scientific information that is pertinent to a series of specific scientific and policy questions suggested to us by leaders in the Southern Commercial Forest, Eastern Spruce-Fir and Eastern Hardwood Research Cooperatives within the Forest Response Program or that we developed ourselves. This document was begun in 1986 and submitted during the last half of 1988. The references and information used in this document, therefore, reflect this time frame. No data from the Forest Response Program were yet available although many research projects were underway.

This document concentrates on the effects of airborne pollutant chemicals on trees and in forest ecosystems of eastern North America by way of foliage-mediated response mechanisms. Many controlled fumigation studies have exposed

seedlings in chambers to determine the exposure-responses of a variety of trees to different pollutant chemicals. With the exception of some of the studies dealing with physiological responses such as photosynthesis and carbon allocation, the authors have based their statements on currently available field data. This document is being published at the present time because it provides a summary of the state of knowledge of the responses of forest trees to aboveground chemical pollutant exposures and the possible forest ecosystem effects resulting from them. This information concerning pollutant effects on the forests of eastern North American has not been previously presented in a single document.

Though this document concentrates on the responses of trees to exposure to airborne chemical pollutants entering through the leaves, trees are also exposed through the soil and roots. Exposures to acid deposition and its sulfate and nitrate components via soil processes are discusssed in this document only briefly. Binkley and others (1989) have discussed in detail the impact of acid deposition on forest soils in the southeastern United States.

Whenever possible, the responses to each question have been formulated as simple statments that concisely present the scientific understanding about the question. Each statement is followed by references from the peer-reviewed scientific literature from which the statement was derived, except in those few instances where such peer-reviewed publications were not available. The authors have sought critical review of this document and each of its component sections by scientists whose knowledge and expertise is greater than our own. Our hope is that this document will provide an understanding of the probable role of acid deposition, photochemical oxidants, and other airborne sulfur- and nitrogen-derived pollutant chemicals in the growth and development of forests in eastern North America and will be of assistance to the Forest Response Program and other divisions of the National Acidic Precipitation Assessment Porgram who are currenty developing a comprehensive assessment of air pollutant effects on forest ecosystems.

Airborne Chemicals of Interest

The airborne chemicals addressed in this document are those known to be or presumed to be injurious to forest trees or ecosystems. Primary air pollutant chemicals are emitted as gases or other volatile waste products from mobile or stationary sources directly into the atmosphere where they may be subsequently transformed chemically into secondary pollutants. Ecosystems may be exposed to either primary or secondary pollutants or both when deposited from the atmosphere as gases and aerosols, and as both dissolved and suspended substances in precipitation, fog, and cloud water. Determination of ecosystem exposure and response, therefore, requires knowledge of the source, chemical form, transport and deposition of the primary airborne pollutants emitted into the atmosphere. When primary pollutants, are converted into secondary pollutants, knowledge of their transport and deposition also is required. Concentration, residence time in the atmosphere and time of year the exposures occur are also important when determining the effects of ecosystem exposure. The airborne chemicals addressed in this document include gases, aerosols, and both dissolved and suspended substances in precipitation. The specific compounds of interest include:

- gaseous sulfur dioxide, nitric oxide, and nitrogen dioxide;
- aerosols and precipitation containing sulfate, nitrate, ammonium, and hydrogen ions;
- gaseous ammonia and nitric acid vapor;
- gaseous ozone, hydrogen peroxide, peroxyacetyl nitrate (PAN), peroxypropionyl nitrate (PPN), and other photochemical oxidants.

Organization of the Document

This paper is organized in four distinct chapters:

Chapter 1 -- Executive Summary

Chapter 2 -- Emissions of Airborne Sulfur- and Nitrogen-Derived Chemicals over North America

Chapter 3 -- Chemical Exposure of Forests in Eastern North America

Chapter 4 -- Effects of Acid Deposition, Photochemical Oxidants, and Other Airborne Sulfur- and Nitrogen-Derived Pollutant Chemicals on Forest Ecosystems.

Chapters 2 to 4 are organized around the series of specific scientific questions about the role of acid deposition, photochemical oxidants, and other airborne sulfur- and nitrogen-derived pollutant chemicals in the forests of eastern North America. These questions are designed to show the possible relationship between the increasing emissions of airborne pollutant chemicals and the effects observed in the forest ecosystems.

These questions are covered in Chapter 2:

- What sulfur- and nitrogen-containing compounds are emitted into the atmosphere of eastern North America?
- What are the principal natural and man-made sources of these compounds?
- In what amounts and chemical and physical forms are these compounds emitted in various regions within North America?
- What temporal changes in emissions of sulfur- and nitrogen-derived compounds have occurred during the industrialization and electrification of various regions within eastern North America?
- In what directions and over what distances are major airborne nitrogen and sulfur compounds usually dispersed from their sources?

- What chemical and physical transformations of sulfur and nitrogen compounds and volatile organic compounds occur during transport from natural and man-made sources and deposition into forest ecosystems?

These questions are covered in Chapter 3:

- What is the chemical exposure of forests in various regions of eastern North America?
- In what chemical and physical forms and by what physical processes are sulfur- and nitrogen-derived pollutants deposited into the forests of eastern North America?
- What is the extent of geographical and elevational variation in deposition of these compounds in various types of forests across eastern North America?
- In what concentrations and during what times of day and seasons of the year are these compounds deposited in forest ecosystems?

These questions are examined in Chapter 4:

- How do forest ecosystems respond to changes in the physical and chemical climate?
- By what physical, chemical, and biological processes are these compounds taken up by plants, and microorganisms in forest ecosystems, and what interactions exist among them?
- What is the likelihood that acid deposition and other airborne sulfur- and nitrogen-derived chemicals affect the forests of eastern North America through foliage-mediated response mechanisms?
- What is the likelihood that airborne chemical pollutants affect forest ecosystems through soil-mediated response mechanisms?
- What effects are airborne chemical pollutant stresses having on forest ecosystems of eastern North America?

1. EXECUTIVE SUMMARY

1.1 Introduction

Natural forest ecosystems in the United States have been exposed to to airborne pollutants for several decades as industrialization, electrification, urbanization and automotive transportation have increased. The chemical pollutant emissions associated with these sources have increased as the number of sources increased. The effects on forest trees of exposures to the chemical emissions have not been evaluated or quantified except in Southern California and on white pine in the East. Changes in the condition of forests in various parts of eastern North America, however, have been observed. Decreased growth, dieback, and mortality of certain tree species have been reported for high-elevation forest ecosystems from Maine, New Hampshire, Vermont and New York south to North Carolina and Tennessee. Decreased growth in natural stands of yellow pines in the Piedmont of Georgia, and of North and South Carolina has also been reported. Dendroecological studies suggest that the decreases in growth of forest trees in the eastern United States began during the 1950's or early 1960's. The extent of forest decline and the factors that precipitated them, however, are questions of controversy. The problem is determining causation. Many hypotheses have been advanced as to the possible cause or causes (see table 1 and chapter 4).

Documentation of effects on forest trees is lacking particularly because their size and perennial nature are not conducive to short-term manipulative experiments that make inference of a causal relationship possible. A causal relationship may be inferred when there is a strong pattern of consistency, responsiveness, and a proven biological mechanism. Any two, of the above three linkage patterns may be sufficient to infer cause in simple systems, however, more complicated systems may require all three. To determine consistency, the symptoms of injury or disfunction must be associated consistently with the presence of the causal factor. To determine mechanism, the suspected cause must be related to an observed effect through one or more biological processes. Responsiveness is the most difficult to prove in mature ecosystems as it requires development of an exposure-response relationship based on the exposure of healthy trees to various known concentrations of airborne pollutants under controlled conditions that simulate those in the forest (see chapter 4).

Responses of seedlings exposed to known concentrations of air pollutants under controlled conditions cannot be readily extrapolated to mature trees (see chapter 4). Responses of mature forest trees to stresses resulting from exposure to ambient air pollutants are difficult to determine because they are usually subtle growth responses that are the cumulative result of many years of multiple exposures. Altered growth dynamics of trees over time may, depending on the severity of the response (e.g., mortality, loss in competitiveness), bring about changes in forest community growth and development. Such stress responses can also be expected to be strongly influenced by site variables, such as soil chemistry, the developmental state of the stand, and by climatic factors such as temperature and rainfall. These factors have made it

Table 1. Some Hypotheses to Explain Forest Decline in Eastern North America

Category	Hypothesis
Natural process	Synchronous canopy dieback Biotic pathogens Assorted abiotic stress agents
Forest stand history	Landscape conversion Forest management practices (e.g., genetic stock) Forest fire frequency and intensity
Anthropogenic stress	Nitrogen subsidy (e.g., nitric acid vapor, nitrate deposition) Direct ozone effects (e.g., inhibition of photosynthesis) Indirect ozone effects (e.g., leaching) Direct effects of hydrogen ion deposition Indirect effect of hydrogen ion deposition (e.g., rhizosphere aluminum toxicity) Deposition of sulfur Deposition of organics Deposition of multiple pollutants (wet and dry deposition) Deposition of trace elements Acid cloud moisture
Multiple factors	Site-specific combination of assorted natural processes, forest stand history, and anthropogenic stress

Adapted from Taylor and Norby (1985).

difficult to determine the effects of air pollutants on forest trees and on the ecosystems of which they are a part (see chapter 4).

The following information explains the purpose of this document and presents the analyses and conclusions of the authors based on the data available.

1.2 Purpose of This Document

This document was developed to help resolve the apparent conflict between:

a) The prevailing scientific judgment that ozone and other photochemical oxidants, either alone or in combination with acid deposition, are the airborne pollutant chemicals most likely to adversely affect southern commercial forests, eastern hardwood forests, and possibly eastern spruce-fir forests;

b) the prevailing public perception that emissions of sulfur and nitrogen oxides leading to increased deposition of acidic and acidifying substances are the airborne chemicals most likely to adversely affect these same forests.

1.3 Airborne Chemicals of Interest

Forest ecosystem responses to stresses, because they are the summation of multiple responses of individual organisms and their interactions, usually require at least a generation (30-50 years) before their expression can be observed unless they are the result of a catastrophe. Therefore, determining the effects of airborne pollutant chemicals on forest ecosystems requires a knowledge of the sources, chemical composition and volume of emissions and whether these have changed over time. It also requires an understanding of the transformation, transport, atmospheric duration, deposition and uptake phenomena associated with the movement of airborne chemical pollutants into leaves of forest trees. Meteorological features of the terrain on which forests grow and time of year when exposures occur also are important factors when determining responses.

For these reasons, the airborne chemicals addressed in this document include the primary and secondary pollutants that are transferred from the atmosphere into forest ecosystems in tne form of gases and aerosols (dry deposition), and both dissolved and suspended substances in precipitation, fog and cloud water (wet deposition). Primary pollutants are gases or other volatile waste products emitted directly from stationary or mobile sources. Secondary pollutants are formed in the atmosphere by chemical transformation of primary pollutants.

The three most important primary air pollutants are sulfur dioxide (SO_2), nitrogen oxides (NO_2 and NO), and volatile organic compounds (VOC). Smaller amounts of gaseous ammonia, sulfuric acid, sulfate aerosol, and hydrochloric acid also are emitted as primary pollutants (see chapter 2).

The most important secondary pollutants include two general classes of compounds -- photochemical oxidants and acid deposition. Photochemical oxidants include ozone (O_3), hydrogen peroxide (H_2O_2), peroxyacetyl nitrate (PAN), and peroxypropionyl nitrate (PPN).

The constituents of acidic and acidifying deposition include

- gaseous sulfur dioxide, nitrogen oxides, nitric acid, and ammonia;
- fine and coarse aqueous aerosols containing sulfuric and nitric acids and all the major nutrient cations and anions including sulfate, nitrate, phosphate, chloride, potassium, sodium, magnesium, ammonium, and hydrogen ions; and
- precipitation and cloud water containing dissolved or suspended matter including all of these major nutrient cations and anions -- sulfate, nitrate, phosphate, chloride, potassium, sodium, magnesium, ammonium, and hydrogen ions (see chapter 2 and chapter 3).

1.4 Airborne Chemical Pollutant Exposure of Forests in Eastern North America

Forests cover more than 50 percent of the total land area of eastern North America. The average distance between major stationary sources and urban centers in the eastern United States is considerably less than the average daily distance (100-200 km) that both primary and secondary pollutants are transported. Thus, most forests in eastern North America (with the exception of the most northerly portions of Ontario and Quebec) are within the area where exposure to either primary and secondary pollutants or both could occur.

Proximity to pollutant sources is particularly important in determining the exposure of forests to sulfur dioxide, nitrogen oxides and pollutant mixtures. Ozone, however, can be transported for hundreds of miles and be enriched as the air masses move over urban and other areas where additional ozone or its precursors are present. Sulfates and nitrates, secondary pollutants formed from SO_2 and NO_2, also may be transported long distances before they are deposited into forest ecosystems.

With the possible exception of ozone and hydrogen peroxide, all of the primary and secondary airborne chemicals of interest in this document, depending on circumstances, can have either stimulatory or inhibitory growth effects on forests:

-- stimulatory effects at low concentrations or loadings; and

-- inhibitory effects at high concentrations or loadings. Unfortunately, the demarcation between growth-stimulating and growth-inhibiting chemical exposures is not known for most airborne pollutant chemicals (see chapter 3).

In characterizing the chemical exposure of forests of eastern North America to atmospheric pollutants, we found it useful to recognize five general types that, because they differ appreciably in proximity to major sources of pollutants, or are subject to distinctive chemical meteorology, could differ significantly in terms of pollutant exposure. These types include the following:

1) Urban forests such as the 30,000 hectares of forest land within the city limits of Atlanta, GA. Although urban forests make up only a very small percentage of the total forest area of eastern North America, these areas contain about 30 percent of the people of the United States and Canada and thus are of interest to many citizens in both countries;

2) Rural forests near major point sources (for example, within approximately 30 km of a major power plant or metal smelter). We estimate that these areas make up about 5 percent of the total forest area of the eastern United States and southeastern Canada;

3) Near-urban forests within about 50 km of major area sources of pollution. We estimate that these near-urban areas could include as much as about 50 percent of the total forest area of the eastern United States and southeastern Canada;

4) Rural forests more than about 50 km from major urban areas or other large area sources of pollution. We estimate that these areas make up about 25-30 percent of the total forest area of the eastern United States and southeastern Canada;

5) High elevation forests such as those on Mount Mitchell in North Carolina or Whiteface Mountain in New York (see fig. 3). We estimate that these areas make up less than 1 percent of the total forest area of the eastern United States and southeastern Canada (see chapter 3).

The exposure of high-elevation forests (>1,000 meters above sea level) to pollutant chemicals is quite different from that of low elevation forests. The reasons for this are listed below.

1) Precipitation inputs are much greater than at low elevation;

2) Cloud-water inputs at high elevations significantly augment precipitation inputs;

3) Concentrations of dissolved and suspended substances in fog and cloud water are typically 6-10 times greater than in precipitation;

4) Ozone and hydrogen peroxide concentrations increase directly with altitude. Maximum ozone concentrations observed at elevated mountainous sites, as well as at many non-mountainous rural and remote sites, often occur at night. This is particularly important for species such as eastern white pine whose stomata remain open at night (see chapter 4).

5) Sites at higher elevations are often exposed to sustained or multiple peak concentrations of ozone within a given 24-hour period as a result of conditions such as (a) trapping inversions; (b) the successive transport of ozone from multiple urban sources upwind, either aloft or across terrain devoid of sufficient ozone scavengers; and (c) the occurrence of mountain-valley and upslope-downslope flows, such that the trajectory of an air parcel passes back over the same forest or stands of trees (see chapter 4).

6) Wind speeds are greater than at low elevation and thus augment aerosol impaction rates; and

7) Evaporation of fog and cloud water further increase the concentrations of airborne chemicals to which these forests are exposed (see chapter 3).

Tables 2 and 3 show the range in amounts of the various pollutants to which forests of the five types listed above may be exposed in the southeastern and northeastern States of the United States and southeastern provinces of Canada (see chapter 3).

In developing the data on chemical exposure of forests as summarized in tables 2 and 3 and in drawing certain of the General Conclusions listed later in this Executive Summary, we have found it useful to combine the data on chemical exposure of urban forests and rural forests near major point sources, and also the data on exposure of near urban and rural forests.

In addition, analyses of data on pollutant exposure indicate the following:

1) Gaseous sulfur dioxide rarely occurs in concentrations sufficient to cause visible injury to vegetation (50 to 5,000 ppb for 8 hours or 500 to 3,000 ppb for 1 hour) in remote, rural, or near-urban forests in eastern North America. (The occurrence of visible injury does not necessarily indicate that decreases in growth or significant changes in the health or productivity of whole forest stands will occur). In high-elevation forests sulfur dioxide concentrations are usually near the limits of detection. Injurious concentrations do occur in some urban and in rural forests near major pollution sources. Maximum concentrations of 500 to 1,000 ppb or even higher were measured near industrial sources for several days during the 1970's and early 1980's (see chapter 3).

2) Nitrogen oxides are rarely, if ever, found in concentrations sufficient to cause visible injury to vegetation (2,000 to 5,000 ppb for 8 hours or 4,000 to 8,000 ppb for 1 hour) in eastern North America (see chapter 3). This is true in part because nitrogen oxides are often consumed in photochemical oxidant formation.

3) Ozone occurs in concentrations sufficient to cause visible injury to vegetation in most of eastern North America. For example, episodes lasting from hours to weeks and exposing eastern North America from the Midwest to the Atlantic Coast with concentrations ranging from 60 to a maximum of 300 ppb have been reported. The highest 1-hour peak concentrations of ozone in the eastern United States occur in New England, the mid-Atlantic States and the Gulf Coast. (Peak concentrations usually cause the most severe vegetation injury). From 1981 to 1983, peak concentrations of 170, 180, 250 and 280 ppb were reported for Raleigh, NC; Boston, MA; Newark, NJ; and Houston, TX, respectively (see chapter 3).

4) Co-occurring and sequential exposure to sulfur dioxide, nitrogen oxides, and ozone have been investigated using 30 monitoring sites in the eastern United States. Sulfur dioxide and ozone co-occurred approximately 30 percent of the time. Sulfur dioxide and ozone co-occurred because ozone was frequently

Table 2. Typical range of chemical exposure in forests in the Southeastern United States reported in refereed publications

Pollutant Chemical	Urban Forests & Rural Forests Near Major Sources	Near-Urban & Rural Forests	High-Elevation Forests
Primary Pollutants (1-hr average values):			
Sulfur dioxide (ppb)	<5-800	<5-200	<1-20
Nitrogen oxides (ppb)	<5-200	<5-100	<3-40
Photochemical Oxidants (1-hr average values):			
Ozone (ppb)	<20-250	<20-200	<10-110
Hydrogen peroxide in cloud water (μmol/L)	Unknown	Unknown	<5-60
Nitric acid (ppb)	<1-10	<1-5	<1-3
PAN (ppb)	Not reported	Not reported	Not reported
PPN (ppb)	Not reported	Not reported	Not reported
Acidic and Acidifying Deposition:			
Sulfate ion (kg/ha/yr):			
from sulfur dioxide	Unknown	Unknown	Unknown
in precipitation	10-20	10-20	10-20
in cloud water	--	--	10-15
in sulfate aerosol	10-15	10-20	10-15
Nitrate ion (kg/ha/yr):			
from nitrogen oxides	Unknown	Unknown	Unknown
in precipitation	5-15	5-10	10-40
in cloud water	--	--	5-20
in nitrate aerosol	5-15	5-10	5-10
Ammonium ion (kg/ha/yr):			
from ammonia	Unknown	Unknown	Unknown
in precipitation	1-2	1-3	1-2
in cloud water	--	--	1-2
in ammonium aerosol	1-2	1-2	1-2
Hydrogen ions (g/ha/yr):			
from HNO_3 & HCl	Unknown	Unknown	Unknown
in precipitation	100-200	100-200	300-400
in cloud water	--	--	300-400
in aerosol	100-200	100-200	300-400

Table 3. Typical range of chemical exposure in forests in the Northeastern United States and Southeastern Canada reported in refereed publications

Pollutant Chemical	Urban Forests & Rural Forests Near Major Sources	Near-Urban & Rural Forests	High-Elevation Forests
Primary Pollutants (1-hr average values):			
Sulfur dioxide (ppb)	<5-1,000	<5-210	<1-15
Nitrogen oxides (ppb)	<5-400	<5-300	<3-50
Photochemical Oxidants (1-hr average values):			
Ozone (ppb)	<20-200	<20-150	<10-90
Hydrogen peroxide in cloud water (μmol/L)	Unknown	Unknown	<5-80
Nitric acid (ppb)	<1-10	<1-5	<1-3
PAN (ppb)	Not reported	Not reported	Not reported
PPN (ppb)	Not reported	Not reported	Not reported
Acidic and Acidifying Deposition:			
Sulfate ion (kg/ha/yr):			
from sulfur dioxide	Unknown	Unknown	Unknown
in precipitation	10-35	10-35	10-35
in cloud water	--	--	10-20
in sulfate aerosol	10-35	10-35	10-15
Nitrate ion (kg/ha/yr):			
from nitrogen oxides	Unknown	Unknown	Unknown
in precipitation	10-20	10-15	10-40
in cloud water	--	--	5-25
in nitrate aerosol	5-15	5-10	5-15
Ammonium ion (kg/ha/yr):			
from ammonia	Unknown	Unknown	Unknown
in precipitation	2-5	2-5	1-2
in cloud water	--	--	1-2
in ammonium aerosol	2-5	2-5	1-2
Hydrogen ions (g/ha/yr):			
from HNO_3 & HCl	Unknown	Unknown	Unknown
in precipitation	400-600	400-600	400-600
in cloud water	--	--	300-400
in aerosol	400-600	400-600	400-600

present at concentrations greater than 50 ppb, and concentrations of sulfur dioxide greater than 50 ppb occurred periodically. When SO_2 concentrations were 30 ppb or lower, no co-occurrences were observed (see chapter 3).

5) Exposure of forests or their components to the photochemical oxidants listed below have not been studied. Controlled experiments have shown that most woody plants are not sensitive to PAN.

- Gaseous hydrogen peroxide;
- Hydrogen peroxide dissolved in cloud water and precipitation;
- Nitric acid;
- PAN; and
- PPN.

6) Exposure of forest trees to gasesous chemical pollutant stresses, whether severe acute stresses or long-term chronic stresses occurs chiefly through the stomata, the same openings in leaves, needles or young shoots through which plants exchange gases with the atmosphere during the processes of photosynthesis, respiration and transpiration (see chapter 4).

7) Transfer of gases from the atmosphere to the physiological sites of action within the leaf interior involves both a gas phase and a liquid phase. The processes of deposition are the crucial links between the transport of atmospheric pollutants to forest canopies, their uptake by trees and the manifestation of effects Any factor that influences pollutant deposition will change the relationship between the concentrations of the pollutant in the atmosphere and the corresponding concentration at the sensitive site within the leaf cells (see chapter 4).

8) Gases moving into the leaf encounter concentrations of transpired water vapor and a variety of hydrocarbons, (e.g., terpenoids) that are produced within the leaf in secondary metabolism, volatilized into the gas phase of the leaf interior, and emitted through the stomata. These substance are more highly concentrated within the leaf boundary layer and may significantly affect the surface and uptake of gases and particles (see chapter 4).

9) Stomatal control of diffusion into the leaf is a major factor influencing the exchange of gaseous pollutants between the atmosphere and internal leaf tissues. All studies indicate that stomatal resistance is the most dynamic and most influential resistance to transfer of gaseous ozone, sulfur dioxide and nitrogen oxides when the canopy is dry. Stomatal functioning is controlled by both physiological and bioenvironmental processes (see chapter 4).

1.5 Critical Loadings of Airborne Pollutant Chemicals

Total loading of sulfate, nitrate, ammonium, and hydrogen ions (amount deposited into a forest or aquatic ecosystem) is the result of wet and dry deposition. The effects of these chemicals on vegetation and forest ecosystems, regardless of whether they are added through acid precipitation or in dry form, is chiefly through soil. The soil-mediated responses of forests in the Southeast to acid deposition are discussed in detail by Binkley and others (1989) and are, therefore, not emphasized in this document.

Evidence now accumulating suggests that the addition of nitrate and ammonium to the soil plays a significant role in the growth of forest trees. Concern has arisen regarding the possible effects on both terrestrial and aquatic ecosystems of these additions. Attempts are being made to determine a critical load. A critical load is defined as the highest load that will not cause chemical changes leading to long-term harmful effects on the most sensitive ecosystems (see chapter 4). The definition carries with it the notion that there is a load at which no long-term effects occur. This concept has been used in attempts to determine the effects of sulfate and nitrate deposition on ecosystems. Though the effects of sulfate on aquatic ecosystems have been studied for many years, critical loadings have not been determined for any of the sulfur- or nitrogen-derived constituents of acid deposition. Certain important facts concerning the effects of deposition into forest ecosystems are listed below.

1) Exposure of forests is through wet and dry deposition. In general, sulfate concentrations are greater in summer than in winter. However, in a broad band extending from New England southwestward along both sides of the Applachian Mountains as far western North Carolina, the ratio of wintertime to summertime sulfate concentration is about two to one (see chapter 3).

2) Wet deposition of sulfate and nitrate ions in warm periods is highly episodic. About 50 to 70 percent of the total annual deposition of sulfate and nitrate, occurs in approximately 20 percent of the total days; however, nitrate deposition is not as seasonally dependent.

3) Ambient deposition of nitrogen is much greater in high- than in low- elevation forests, therefore, detrimental effects of total nitrogen are more likely in high-elevation forests (chapter 3).

4) Ambient deposition of airborne sulfur and nitrogen compounds (including gases, aerosols, and dissolved and suspended substances in precipitation and cloud water) provides amounts of nutrient sulfur and nitrogen that probably increase growth of forest trees in rural, near-urban, urban, and both high- and low-elevation forests in some parts of eastern North America. This is true because the total nutrient input from atmospheric sources of the several compounds involved provides an important part (10 to 30 percent) of the annual ecosystem demand for total nutrient sulfur and nitrogen (see chapter 3 and chapter 4).

5) The total supply of airborne nutrient nitrogen includes the following compounds: gaseous nitrogen oxides + gaseous nitric acid, + gaseous ammonia; ammonium + nitrate aerosols; ammonium + nitrate ions in precipitation; and ammonium + nitrate ions in fog and cloud water; and possibly PAN and PPN.

6) The total supply of airborne nutrient sulfur includes the following compounds: gaseous sulfur dioxide + sulfuric-acid and sulfate aerosols + sulfate ions in precipitation and fog and cloud water.

7) Present evidence is not sufficient to accept or reject any of the hypothetical mechanisms for action of acid deposition on forest tree growth by either foliage- or soil-mediated responses. Such evidence does suggest, however, that direct injury to foliar organs by ambient acid deposition is much less likely than indirect effects mediated through interference with normal foliar and root nutrient uptake and leaching processes (see chapter 3 and chapter 4).

1.6 General Conclusions

1. At ambient concentrations and rates of deposition observed in the eastern United States, neither acid deposition in the form of aerosols nor in the form of dissolved or suspended substances in precipitation or cloud water have been shown to induce acute detrimental effects on forests or crops via foliage-mediated response mechanisms. Deposition of sulfate and nitrate onto the soil during acid precipitation, however, can affect tree growth (see chapter 4).

2. The pH of the ambient precipitation (pH 4.0-6.0) in most of the eastern United States is substantially higher than the pH of simulated acid precipitation necessary to induce visible injury to forest trees or commercial crops (pH 2.0-3.5), with the exception of one cultivated variety of soybeans (see chapter 3 and 4).

3. Gases, fine and coarse aerosols (including fog and cloud water droplets), and coarse particulate matter are transferred into forests by dry deposition processes. Dry deposition occurs under all meteorological conditions, at all times of the day and night, and in all seasons of the year. Dry deposition is more difficult to quantify than wet deposition (chapter 3).

4. Toxic gases at ambient concentrations are the only physical forms of airborne pollutant chemicals known to cause visible symptoms of injury, decreased growth, or mortality of forest trees. Toxic gases enter plants mainly through leaf stomata.

5. In eastern North America, sulfur dioxide rarely occurs in high enough concentrations (above 50 ppb for 8 hours) to be injurious to forests on a broad regional scale. In addition, in northern

areas where residential and commercial space and water heating are major sources, sulfur dioxide concentrations are usually higher during the winter months when most trees are not actively growing.

6. Sulfur dioxide injury to vegetation is found most frequently in the vicinity of strong point sources such as metal smelters and power plants. Vegetation exposure to phytotoxic concentrations in these areas is usually of short duration. Exceptions are Wawa and Sudbury, Ontario, Trail, British Columbia, and Copper Hill Basin near Ducktown, TN, where catastrophic emissions resulted in long-term injury. (see chapters 3 and 4).

7. Based on long-term trends in emissions, it may be inferred that sulfur (sulfur dioxide and sulfate) loadings of forest ecosystems have remained relatively constant or decreased in the northeastern United States and southeastern Canada since the 1920's, but have increased steadily in the southeastern United States since 1940's (see chapter 3).

8. Gaseous nitrogen oxides at ambient concentrations (50 ppb) have not been proven to cause visible injury, decreased growth, or mortality of either forest trees or agricultural crops at any location in North America (see chapter 3 and 4).

9. Gaseous ozone occurs in eastern North America in concentrations sufficient to cause foliar injury, inhibit photosynthesis, alter carbon allocation, and in tree roots, interfere with mycorrhizal formation. Disruption of these important physiological and biochemical processes can suppress the growth of trees, shrubs, and herbaceous vegetation by decreasing their capacity to form carbon compounds and their ability to absorb water and needed nutrients from the soil. Ozone has been shown to cause foliar injury, decreased photosynthesis and reduced growth in a number conifer and hardwood species (see chapter 4).

10. Ambient ozone concentrations between 40 and 70 ppb, or higher, for 1 to 7 days during the growing season have reduced the growth of sensitive vegetation, intermediately-sensitive vegetation and some tolerant vegetation. Growth of sensitive tree species has been reduced without causing foliar injury. Present evidence is not sufficient, however, to determine if ozone is causing economically significant injury to the health or productivity of whole stands of eastern white pine, other eastern conifers or hardwoods (see chapter 4).

11. Emission trends data indicate a sharp increase for SOx and NOx beginning in the late 1940's. Ozone concentrations, fueled by large-scale NOx emissions that began during World War II, undoubtably also increased in the late 1940's even though data indicating this does not exist. Circumstantial evidence suggests that the ecosystems of eastern North America have been subject to phytotoxic episodic ozone concentrations, and sulfate and nitrate deposition, for at least 30, possibly 40 years (see chapter 3 and 4).

12. Field studies and observations support the view that ozone has been a important gaseous pollutant in the eastern United States for many years. Vegetation injury, resulting ozone exposure was first observed in New Jersey on many cultivated crops, ornamentals and indigenous vegetation beginning in 1944, though the cause of the injury was not known until 1950 when Heggestad and Middleton reported ozone injury of tobacco (see chapter 4).

13. In the 1950's, the United States Forest Service reported oxidant-induced injury of eastern white pine on the Cumberland Plateau of East Tennessee. However, at that time the injury was attributed to unidentified atmospheric constituents. Needle blight (also called emergence tipburn) of eastern white pine, the result of acute and chronic oxidant exposure, was reported from West Virginia and western North Carolina in the late 1950's and early 1960's (see chapter 4).

14. Data from studies made since 1976 indicate that ozone episodes in eastern North America are a common phenomenon. Typical ozone episodes are associated with high pressure systems that originate in Canada and move south into the Midwest enroute to the Atlantic Ocean and northward into New England. Other episodes that affected the entire southeastern United States from western Texas northeastward through Illinois, and finally east to the Atlantic Ocean have been associated with high pressure systems over the Gulf of Mexico. In addition, some episodes originating over New York City and moving westward to Washington, DC, and Philadelphia to Pittsburgh and south along the Appalachian Mountains to the Gulf Coast of Florida have been associated with high pressure systems over the Atlantic Ocean (see chapter 3 and 4).

15. Ozone is ephemeral. Unlike residuals of sulfur or nitrogen oxides, fluorides, heavy metals, or radio-nuclides, ozone or its by-products, are not selectively retained or accumulated in any particular components of forest ecosystems (see chapter 4).

16. Ozone is a relatively homogenous regional pollutant, (chiefly due to the sources of its precursors and the processes that contribute to its formation). It is the one gaseous airborne chemical which is capable of having exposed to phytotoxic concentrations without leaving a permanent trace the entire eastern United States where visible injury and growth reductions of forest trees, crops and other vegetation have been observed. It is probably, therefore, that the multiple episodic ozone exposures have been a source of major above-ground stresses on forests and other vegetation for many years (see chapter 3 and 4).

17. Experiments using sulfuric acid aerosols suggest that exposure to acid aerosols may predispose trees to injury by ozone (see chapter 4).

18. Mixtures of toxic gases, fine and coarse aerosols, and coarse particulate matter are known to occur occasionally in the atmosphere near major point and area sources of pollution -- in or near cities or near power plants or metal smelters. But actual field measurements indicate that co-occurrence of phytotoxic concentrations of gaseous ozone, sulfur dioxide, or nitrogen oxides is rare. Thus though present evidence is far from conclusive, the risks to forests in eastern North America from mixtures of gaseous pollutants and aerosols does not appear to be severe (see chapter 3 and 4).

19. Hydrogen peroxide has been detected in cloud water collected in high-elevation forests in eastern North America in concentrations that were sufficient to experimentally cause injury to spruce seedlings in The Netherlands. Attempts in North American to demonstrated similar injurious effects at these same concentrations on spruce and pine seedlings have failed to confirm the observations made in The Netherlands (chapter 3 and 4).

20. Tree growth is the culmination of many biochemical and physiological processes. Virtually every perturbation of a plant community results in stress and affects the performance and surivival of individual plants. The age of the tree, the complex interactions among pollutant and natural stresses combined with various environmental and genetic factors and the action these have on physiological and biochemical processes within the tree, determine the impact that ambient chemical pollutants have on growth (see chapter 4).

21. Trees are perennial plants. Thus, they must cope with the cumulative effects of both short- and long-term stresses encountered year after year. They may respond to some stresses rapidly as, for example, when needles of eastern white pine exhibit visible injury symptoms within days of exposure to acute ozone concentrations. In many cases, however, response to stresses is through differential growth, resulting from changes in carbon allocation over many years. Therefore, attributing growth reductions observed today to air pollution exposures that began 30-50 years ago is difficult, because long-term data indicating the presence of phytotoxic concentrations of airborne chemical pollutants is lacking (see chapter 3 and 4).

22. Tree vigor is extremely important in determining the response of trees to insects and pathogens. Sulfur dioxide and ozone, by inhibiting photosynthesis and altering carbon allocation, can predispose to attack by insects (especially bark beetles) and various fungal pathogens (especially root rotting fungi).

23. Changes within forest ecosystems result from altered energy flow and nutrient cycling within the forest communities that compose them. These alterations begin with the responses of individual trees to stresses. Stresses sufficient to cause mortality, to

severely alter growth patterns, or to change the ability of trees to compete can set the stage for ecosystem changes especially if they lead to changes in species composition (see chapter 4).

24. Mature forest ecosystems maintain themselves in an oscillating steady state. Trees are continuously undergoing severe competition for space, light, water, and mineral nutrients. Those trees most susceptible to natural stresses, are eliminated more rapidly by additional and severe periodic stresses such as drought, flooding, fire and attacks by insects or disease-causing organisms. Continuous elimination of old trees and those susceptible to stresses ultimately produces a mature forest with trees and other plant species capable of tolerating the competitional stresses. This usually results in changes in species composition of the ecosystem over time. Unless they are catastrophic (e.g., fire, flood, or windstorm), the effects of natural stresses upon ecosystems are frequently difficult to determine (see chapter 4).

2. EMISSIONS OF AIRBORNE SULFUR- AND NITROGEN-DERIVED COMPOUNDS OVER EASTERN NORTH AMERICA

2.1 Introduction

An understanding of the sources, composition, amounts, and fate of the chemicals being emitted into the atmosphere and how these have changed over the years is essential when quantifying the effects of pollutant exposures on forest ecosystems. The airborne emissions of concern with regard to effects on forests include two major types of air pollutants--primary and secondary. Primary air pollutants are gases or other volatile waste products emitted directly from stationary or mobile sources. Stationary sources include power plants, metal smelters, and other industrial and commercial installations; municipal and industrial incinerators; farms and forests where open burning or volatilization of waste or fertilizer materials is common; domestic and commercial space and water heating units; and municipal and rural sanitary and solid waste disposal systems. Mobile sources are mainly cars, trucks, tractors, trains, aircraft, and ships. Secondary pollutants include a wide variety of substances formed in the atmosphere by chemical transformation of primary pollutants (see fig. 1).

The most important primary ambient air emissions are sulfur dioxide (SO_2) and nitrogen oxides (NO_2 and NO). Smaller amounts of gaseous ammonia, sulfuric acid, sulfate aerosol, and hydrochloric acid are also primary pollutants. The residence time of sulfur dioxide in the atmosphere is 0 to 5 days and of nitrogen oxides is 0 to 4 days. These intervals provide ample time for chemical transformation and dispersion of both primary and secondary pollutants over both short (<500 km) and long distances (>500 km) (National Research Council 1983).

The most important secondary pollutants include two general classes of compounds--photochemical oxidants and acid deposition. The most important photochemical oxidants include ozone (O_3), hydrogen peroxide (H_2O_2), formaldehyde (H_2CO), peroxyacetyl nitrate (PAN), and peroxypropionyl nitrate (PPN). The most important constituents of acid deposition include 1) gaseous sulfur dioxide, nitrogen oxides, nitric acid, ammonia vapor, PAN, and PPN; 2) dry or aqueous aerosols containing sulfate, sulfuric acid, nitrate, ammonium, and other major cations and anions; and 3) all the major cations and anions dissolved or suspended in precipitation and cloud water including sulfate, nitrate, phosphate, chloride, potassium, sodium, magnesium, ammonium, and hydrogen ions (National Research Council 1981).

The average distance between major stationary sources and urban centers in the Eastern United States and southeastern Canada is considerably less than the average daily distance of transport of both primary and secondary pollutants. Thus, most forests in eastern North America (with the exception of the most northerly portions of Ontario and Quebec) are within the zone of influence (deposition field) of primary and secondary pollutants derived from multiple sources of emissions of sulfur dioxide and nitrogen-oxides (National Research Council 1983; U.S. Environmental Protection Agency 1982a, 1982b).

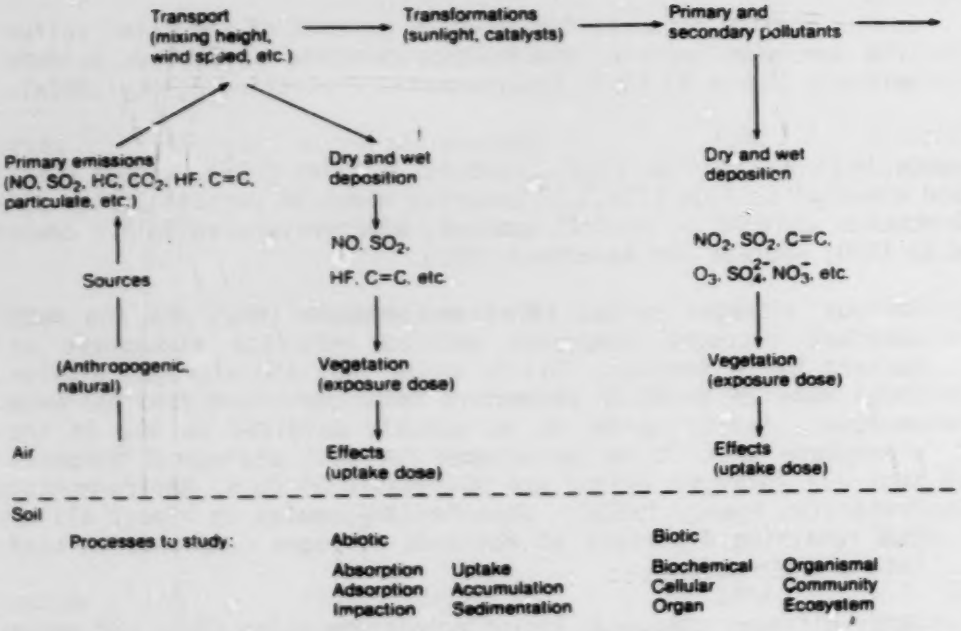


Figure 1. The emission, transport, transformation, and deposition subsystems. The atmospheric portion of biogeochemical cycling. (c = c is ethylene)
Source: Heck (1982).

2.2 What Sulfur- and Nitrogen-Containing Compounds are Emitted into the Atmosphere of Eastern North America?

1) Gasecus sulfur dioxide and nitrogen oxides are the most abundant sulfur and nitrogen compounds emitted into the atmosphere of eastern North America from primary sources (tables 4 and 5) (U.S. Environmental Protection Agency 1982a, 1982b).

2) Sulfur dioxide accounts for about 90 percent of the total sulfur from man-made sources; the balance consists of various sulfate aerosols (table 4) (U.S. Environmental Protection Agency 1982a).

Gaseous hydrogen sulfide (H_2S), carbonyl sulfide (COS), carbon disulfide (CS_2), and dimethyl sulfide [$(CH_3)_2S$] comprise about 98 percent of the airborne sulfur compounds emitted by natural sources; all are reduced sulfur compounds (Sze and Ko 1980; Andreae and Raemdonck 1983).

3) Gaseous nitrogen oxides (NOx) and ammonia (NH_3) are the most abundant nitrogen compounds emitted into the atmosphere of eastern North America. Nitric oxide (NO) and nitrogen dioxide (NO_2) make up about 96 percent of total emissions from man-made sources. Nitric oxide is so quickly oxidized to NO_2 in the atmosphere that it can be assumed for most biological purposes that all nitrogen oxides are NO_2 (table 5) (U.S. Environmental Protection Agency 1982b). Ammonia (NH_3) makes up almost all of the remaining 4 percent of man-made nitrogen compounds emitted into the atmosphere.

   Other nitrogen compounds including nitrous oxide (N_2O) and amines ($-NH_2$) are emitted by man-made sources, but in relatively smaller amounts (U.S. Environmental Protection Agency 1982b).

Nitrous oxide, nitric oxide, and ammonia comprise almost all the airborne nitrogen compounds emitted by natural sources (U.S. Environmental Protection Agency 1982b).

2.3 What are the Principal Natural and Man-made Sources of These Compounds?

1) Sulfur compounds:

   a) The major natural source of airborne SO_2 (98 percent) in eastern North America is the atmospheric oxidation of reduced sulfur compounds. The reduced sulfur compounds of greatest importance are hydrogen sulfide (H_2S), carbonyl sulfide (COS), dimethyl sulfide [$(CH_3)_2S$], dimethyl disulfide [$(CH_3)_2S_2$], and carbon disulfide (CS_2) (table 4). These compounds are found in sea spray and are also produced by decomposition of organic matter by soil and

Table 4. Airborne sulfur compounds and their sources in eastern North America

Compound	Formula	Source	Date	Reference
Hydrogen sulfide	H_2S	biogenic via	1981	Aneja and others 1982 Altshuller and Linthurst 1984
Dimethyl sulfide	$(CH_3)_2S$	microbial process	1981	Altshuller and Linthurst 1984
Dimethyl sulfide	$(CH_3)_2S$	microbial process	1982	Altshuller and Linthurst 1984
Carbonyl sulfide	COS	microbial process	1982	Altshuller and Linthurst 1984
Carbon disulfide	CS_2	microbial process	1982	Altshuller and Linthurst 1984
Methyl mercaptan	CH_3SH	microbial process	1982	Altshuller and Linthurst 1984
Sulfur oxides: (gas phase)	SO_x	fossil fuel combustion	1982	U.S. Environmental Protection Agency 1982a
Sulfur monoxide	SO	fossil fuel combustion	1982	U.S. Environmental Protection Agency 1982a
Sulfur dioxide	SO_2	fossil fuel combustion	1982	U.S. Environmental Protection Agency 1982a
Sulfur trioxide	SO_3	fossil fuel combustion and atmospheric reactions	1982	U.S. Environmental Protection Agency 1982a
Disulfur monoxide	S_2O	fossil fuel combustion	1982	U.S. Environmental Protection Agency 1982a
Sulfate (primary)[a]	SO_4^{2-}	fossil fuel combustion	1984	Altshuller and Linthurst 1984

[a]Nearly 100 percent of aerosol sulfur is in the form of sulfate.

Table 5. Nitrogen-containing compounds and their sources

Compound	Formula	Source	Date	Reference
Nitrogen oxides	NO_x	biogenic; anthropogenic		
Nitric oxide	NO	small amounts of microbial dentrification	1976	Soderlund & Svenson 1976 Altshuller and Linthurst 1984
		fossil fuel combustion	1946	U.S. Environmental Protection Agency 1982b
Nitrogen dioxide	NO_2	fossil fuel combustion; and oxidation of NO	1982	U.S. Environmental Protection Agency 1982b
Nitrous oxide	N_2O	mainly microbial denitrification but some microbial nitrification	1972	Alexander 1977 U.S. Environmental Protection Agency 1982b
Ammonia	NH_3	microbial and insect associated microbial deamination and ammonification; fertilizers; and feedlots	1982	U.S. Environmental Protection Agency 1982b Altshuller and Linthurst 1984
Ammonium	NH_4^+	biogenic; atmospheric	1982	Altshuller and Linthurst 1984 U.S. Environmental Protection Agency 1982b

aquatic microorganisms. These sources produce about 4 percent of the total sulfur emissions for eastern North America (Galloway and Whelpdale 1980).

b) The major man-made sources of airborne sulfur oxides are combustion of coal (65 percent), oil (15 percent), natural gas (<1 percent); the remaining 20 percent are from metal smelters, commercial and residential space, water, and materials heating processes, and industrial processing of metals, wood pulp, etc. (Husar and Patterson 1980).

c) Man-made emissions of sulfur compounds are about 20 times larger than emissions from natural sources in eastern North America. Gaseous sulfur dioxide accounts for more than 90 percent of total man-made sulfur emissions; primary sulfates account for most of the remaining 10 percent (Galloway and Whelpdale 1980).

d) Primary sulfate ($SO_4^=$) is emitted as an aerosol from oil and coal combustion. Emissions of primary sulfate from combustion of oil are 5 to 10 times greater than those from combustion of coal with a similar sulfur content (Altshuller and Linthurst 1984).

2) Nitrogen compounds:

a) Soil microorganisms are the major source of natural airborne nitrogen compounds -- mainly nitrous oxide and NO, while microorganisms and wild animals and insects are the major natural sources of ammonia (NH_3) (U.S. Environmental Protection Agency 1982b). In the Eastern United States, natural sources of nitrogen compounds contribute about one-tenth as much as man-made sources (Soderlund and Svenson 1976).

b) The major man-made sources of airborne nitrogen oxides (mainly NO_2) are combustion of fossil fuels (chiefly coal, oil, gasoline, and natural gas) in mobile (49 percent) and stationary (51 percent) sources (U.S. Environmental Protection Agency 1982b).

c) The major man-made sources of airborne ammonia (NH_3) and amines ($-NH_2$) are volatilization of animal wastes from feedlots (62 percent), fossil fuel combustion (21 percent), ammonia-based fertilizers such as anhydrous ammonia from ammonia-production facilities and farm land after application to soils (12 percent), and industrial sources (5 percent) (U.S. Environmental Protection Agency 1982b). Curiously, volatilization of human wastes was not included as an important source of gaseous ammonia in the inventories reported either by U.S. Environmental Protection Agency (1982b) or National Research Council (1977a). We believe that human wastes must be a significant part of the

total of all volatile sources of nitrogen compounds in eastern North America but have been unable to locate published data either to support or refute this judgment.

d) Man-made emissions of nitrogen compounds are about 10 times larger than emissions from all natural sources in eastern North America (U.S. Environmental Protection Agency 1982b).

2.4 In What Amounts and in What Chemical and Physical Forms are These Compounds Emitted in the Various Regions within North America?

1) Each year about 25 million tons of gaseous sulfur dioxide, about 23 million tons of gaseous nitrogen oxides, and about 3.7 million tons of gaseous ammonia are emitted from man-made sources into the atmosphere over North America (Husar 1986; Husar and Holloway 1983; Husar and Patterson 1980).

2) About 75 percent of these emissions occur in the eastern and only about 25 percent occur in the western parts of the continent (using as demarcation a line running from the Texas-Louisiana border to Manitoba in Canada) (Husar and Holloway 1983).

3) The various states of the United States and the provinces of Canada differ markedly in emissions of both sulfur and nitrogen oxides (see figs. 2 and 3) (National Research Council 1983). The total emissions by region for the year 1980 are shown in table 6 (National Research Council 1983).

4) Emissions of sulfur oxides in most regions, States, and provinces of eastern North America range from about one to three times those of nitrogen oxides. In the States of Texas, Louisiana, Vermont, Connecticut, Rhode Island and New Jersey, however, emissions of nitrogen oxides exceed those of sulfur dioxide. But the regions, States, and provinces with high (or low) sulfur dioxide emissions usually are also high (or low) in emissions of nitrogen oxides (compare figs. 2 and 3) (National Research Council 1983).

2.5 What Temporal Changes in Emissions of Sulfur- and Nitrogen-derived Compounds Have Occurred During the Industrialization and Electrification of Various Regions of Eastern North America?

In our analysis of regional emissions trends, we found it convenient to follow Husar's (1986) lead and divide eastern North America into southeastern Canada and the northeastern, upper and lower midwestern, and southern portions of the United States (fig. 4).

1) From 1880 to 1980, total man-made emissions of sulfur dioxide in eastern North America increased from about 2 million to the present 30 million tons per year (fig. 5a), while total man-made emissions of nitrogen oxides increased from about 1 million to the present 23 million tons per year (fig. 6a) (Husar 1986).

Figure 2. Representative values of SO_2 emissions in the United States and Canada in 1980 (thousands of metric tons).

Source: National Research Council (1983).

Figure 3. Representative values of NO_x emissions in the United States and Canada in 1980 (thousands of metric tons).

Source: National Research Council (1983).

Table 6. Distribution of man-made emissions of sulfur and nitrogen compounds in eastern North America

Sulfur Compounds			Nitrogen Compounds		
Source Region	Metric Tons	Percent	Source Region	Metric Tons	Percent
Total Emissions:					
Eastern Canada	3,271	14	Eastern Canada	1,023	7
Northeast U.S.	3,932	16	Northeast U.S.	2,731	17
Midwest U.S.	8,671	36	Midwest U.S.	4,803	30
Southern U.S.	8,319	34	Southern U.S.	7,276	46
TOTAL	24,193	100	TOTAL	15,833	100
Emissions Density (metric tons per hectare):					
Eastern Canada	1.2×10^{-5}		Eastern Canada	0.31×10^{-5}	
Northeast U.S.	8.5×10^{-5}		Northeast U.S.	5.8×10^{-5}	
Midwest U.S.	7.3×10^{-5}		Midwest U.S.	3.9×10^{-5}	
Southern U.S.	5.0×10^{-5}		Southern U.S.	3.5×10^{-5}	

Source: National Research Council (1983).

2) Total man-made emissions of sulfur dioxide in eastern North America rose rapidly between 1880 and 1920, reached about 20 million tons per year in the early 1920's, then increased again about 50 percent, reaching 30 million tons per year by 1970, after which there was about a 15 percent decrease to about 25 million tons per year, due mainly to increased use of low sulfur fuels (see fig. 5a) (Gschwandtner and others 1986; Husar 1986; National Research Council 1986).

3) These several subregions of eastern North America have differed greatly in temporal patterns of change in man-made emissions of sulfur and nitrogen oxides (see figs. 5 and 6, which show emissions of sulfur per se rather than of sulfur dioxide; please note that 1 ton of sulfur equals approximately 2 tons of sulfate).

For example, with regard to sulfur emissions:

a) In southeastern Canada (Region A), emissions of sulfur dioxide rose from negligible in 1880 to about 2 million tons per year in the 1930's, further rose to about 8 million tons per year in the mid 1970's, and decreased to about 4 million tons per year by 1980 (fig. 5b) (Husar 1986).

Figure 4. Regions in eastern North America selected for the study of regional trends in emissions.

Source: Husar (1986).

Region	
A (Eastern Canada):	Eastern Ontario, New Brunswick, Southern Quebec
B (Northeastern U.S):	Connecticut, Delaware, Maine, Maryland, Massachusetts, New Hampshire, New Jersey, New York, Pennsylvania, Rhode Island, Vermont
C (Southeastern U.S.):	Alabama, Arkansas, Florida, Georgia, Kentucky, Louisiana, Mississippi, North Carolina, South Carolina, Tennessee, Virginia, West Virginia
D (Lower Midwest):	Illinois, Indiana, Michigan, Missouri, Ohio
E (Upper Midwest):	Iowa, Minnesota, Wisconsin
F (Western U.S.):	All states in the contiguous U.S. not included in Regions B to E

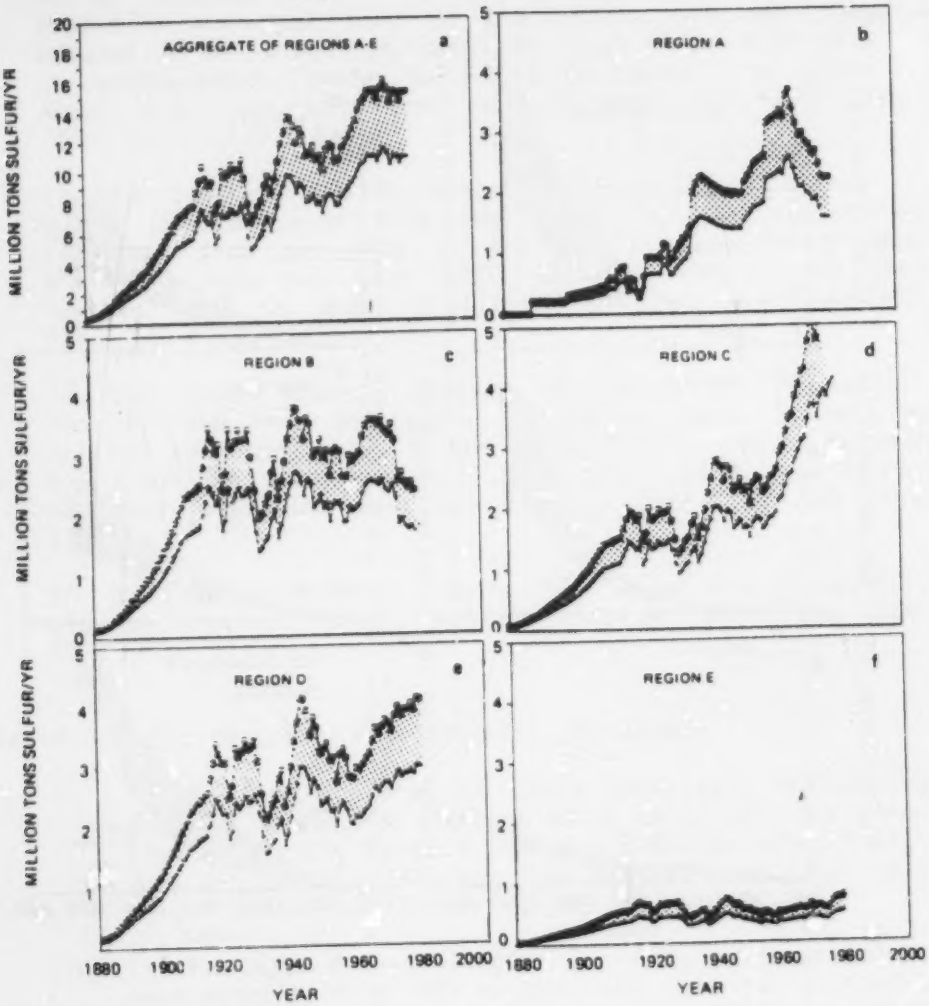


Figure 5. Regional sulfur emission trend estimates for (a) eastern North America (the aggregate of Regions A, B, C, D, and E); (b) Region A; (c) Region B; (d) Region C; (e) Region D; (f) Region E. (SO_2 = tons sulfur multiplied by 2). (Shading indicates range of uncertainty.)

Source: Husar (1986).

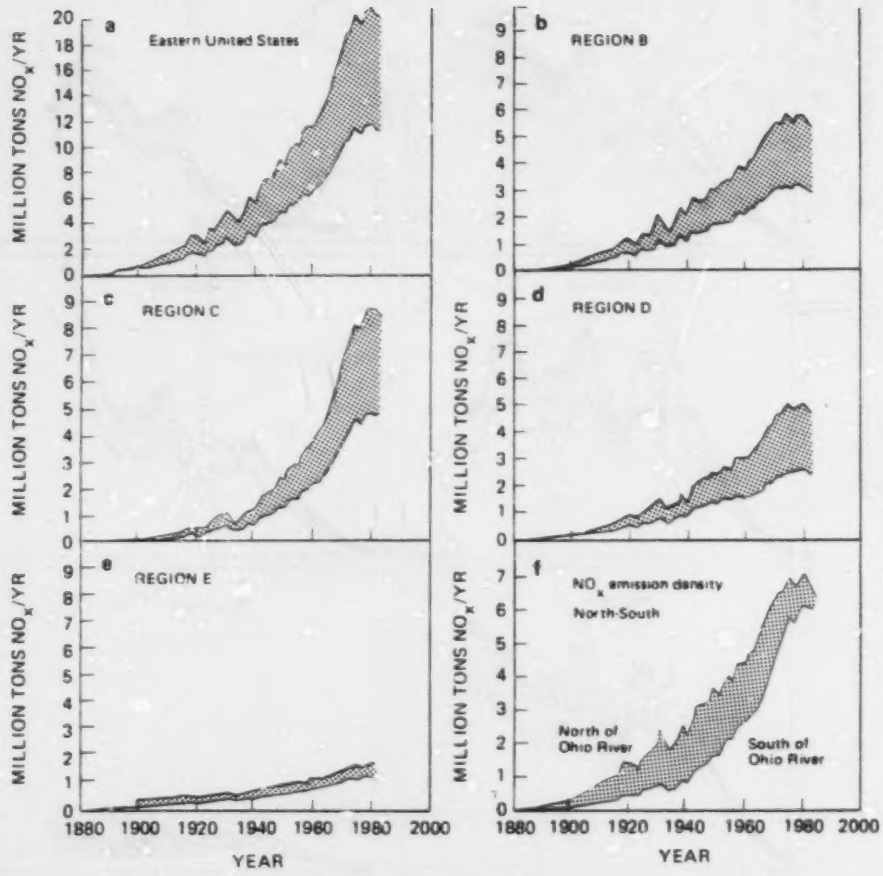


Figure 6. (a) Trends in emissions of nitrogen oxides in the Eastern United States (the aggregate of region B, C, D, and E); (b) Region B: densities of regions north (Regions B, D, and E) and south (Region C) of the Ohio River. (Shading indicates range of uncertainty.)

Source: Husar (1986).

b) In the Northeastern United States (Region B), emissions of sulfur dioxide rose from negligible in 1880 to about 6 million tons per year in the 1920's, and have remained between 4 and 6 million tons per year since that time. As in southeastern Canada, emissions decreased from about 6 to about 4 million tons per year between 1970 and 1980 (fig. 5c) (Husar 1986).

c) In the Southern United States (Region C), emissions of sulfur dioxide rose from negligible in 1880 to nearly 4 million tons per year in the 1920's, remained at about 4 million tons per year until the mid 1960's, and then further rose to about 8 million tons per year by 1980 (fig. 5d) (Husar 1986).

d) In the lower Midwest (Region D), emissions of sulfur dioxide rose from negligible in 1880 to about 3 and a half million tons per year in the early 1920's, varied between 4 and 6 million tons per year until about 1970, and decreased to about 4 million tons per year by 1980 (fig. 5e) (Husar 1986).

e) In the upper Midwest (Region E), emissions of sulfur dioxide remained below 2 million tons per year from 1880 to 1980 (fig. 5f) (Husar 1986).

Similarly, with regard to nitrogen oxide emissions:

a) In the Northeastern and lower Midwestern United States (Regions B and D), nitrogen oxide emissions rose steadily from negligible between 1880 and 1900 to about 4 million tons per year in 1970 and then decreased slightly by 1980 (figs. 6b and 6d) (Husar 1986).

b) In the upper Midwest (Region E), nitrogen oxide emissions rose steadily from 1900 to 1980, but even then reached only about 1 million tons per year (fig. 6e) (Husar 1986).

c) In the Southern United States (Region C), emissions of nitrogen oxides remained below 0.5 million tons per year until the 1920s, but then rose nearly exponentially to 5 to 8 million tons per year by 1980 (fig. 6c) (Husar 1986).

4) In North America as a whole, total man-made emissions of nitrogen oxides rose slowly from 1880 to 1920, reached about 10 million tons per year in the mid 1950's, and 20 million tons per year in the early 1980's (fig. 6a) (Husar 1986). This upward trend is primarily the result of increased use of coal, gasoline, diesel fuel, and natural gas (fig. 7) (Gschwandtner and others 1986).

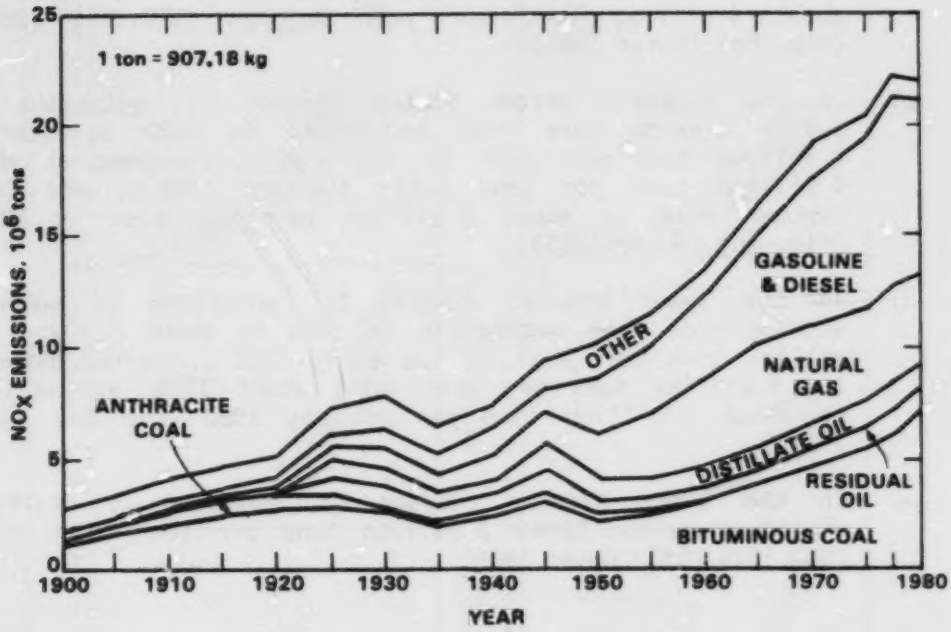


Figure 7. Overall trend in NO_x emissions from 1900 to 1980 for the United States and by fuel type for each study year.

Source: Gschwandtner and others (1986).

5) The increase in total SO_2 emissions caused by the electrification of North American society between 1940 and 1980 is about four times greater than that caused by the industrialization of North American society between 1880 and 1980 (Altshuller and Linthurst 1984). In fact, industrial emissions of sulfur dioxide in the United States decreased from about 7 million tons per year in the late 1920's to about 4 million tons per year in the mid 1950's to 1980; by contrast, electric utility emissions increased from about 3 million tons per year in the early 1940's to about 18 million tons per year since 1970 (fig. 8) (Gschwandtner and others 1986; Husar and Holloway 1983).

6) Total man-made emissions of sulfur dioxide in North America are predicted to remain between 24 and 30 million tons per year for the next 50 years (fig. 9) (U.S. Congress 1984; U.S. Environmental Protection Agency 1984).

7) Total man-made emissions of nitrogen oxides in North America are predicted to continue to increase from about 20 million tons per year to between 22 and 32 million tons per year during the next 50 years (fig. 9) (U.S. Congress 1984; U.S. Environmental Protection Agency 1984).

2.6 In What Directions and Over What Distances are Major Airborne Nitrogen and Sulfur Compounds Usually Dispersed from Their Sources?

1) Once emitted, sulfur and nitrogen compounds are carried wherever the wind blows (National Research Council 1983, 1986).

2) The average zone of influence of any particular emission source extends in all directions from the source, and is roughly symmetrical--with the center of the deposition field generally displaced from the source by a distance of only a few tens or perhaps hundreds of km. Thus, air pollutants generally have their greatest effects within a few hundred km from their source of emission (Bolin and others 1971; National Research Council 1983, 1986).

3) The concentration and deposition of primary air pollutants decreases progressively with increasing distance and time after emission from any source (Altshuller and Linthurst 1984).

4) The concentration and deposition of secondary pollutants is a complex function of meteorological, seasonal, altitudinal, temporal, geographical, and other factors (National Research Council 1983).

5) The elapsed time between emission and deposition varies with the airborne pollutant chemical in question but ordinarily ranges from a few minutes or hours to a maximum of about 5 days for

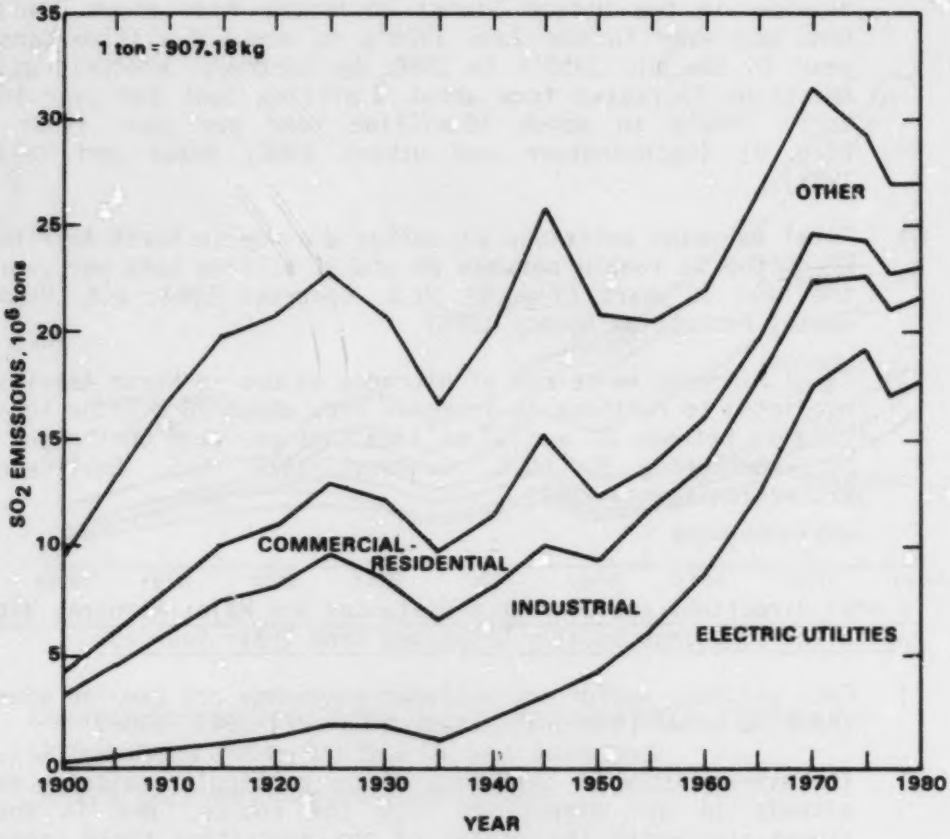


Figure 8. Overall trend in SO_2 emissions from 1900 to 1980 for the United States and by source category for each study year.

Source: Gschwandtner and others (1986).

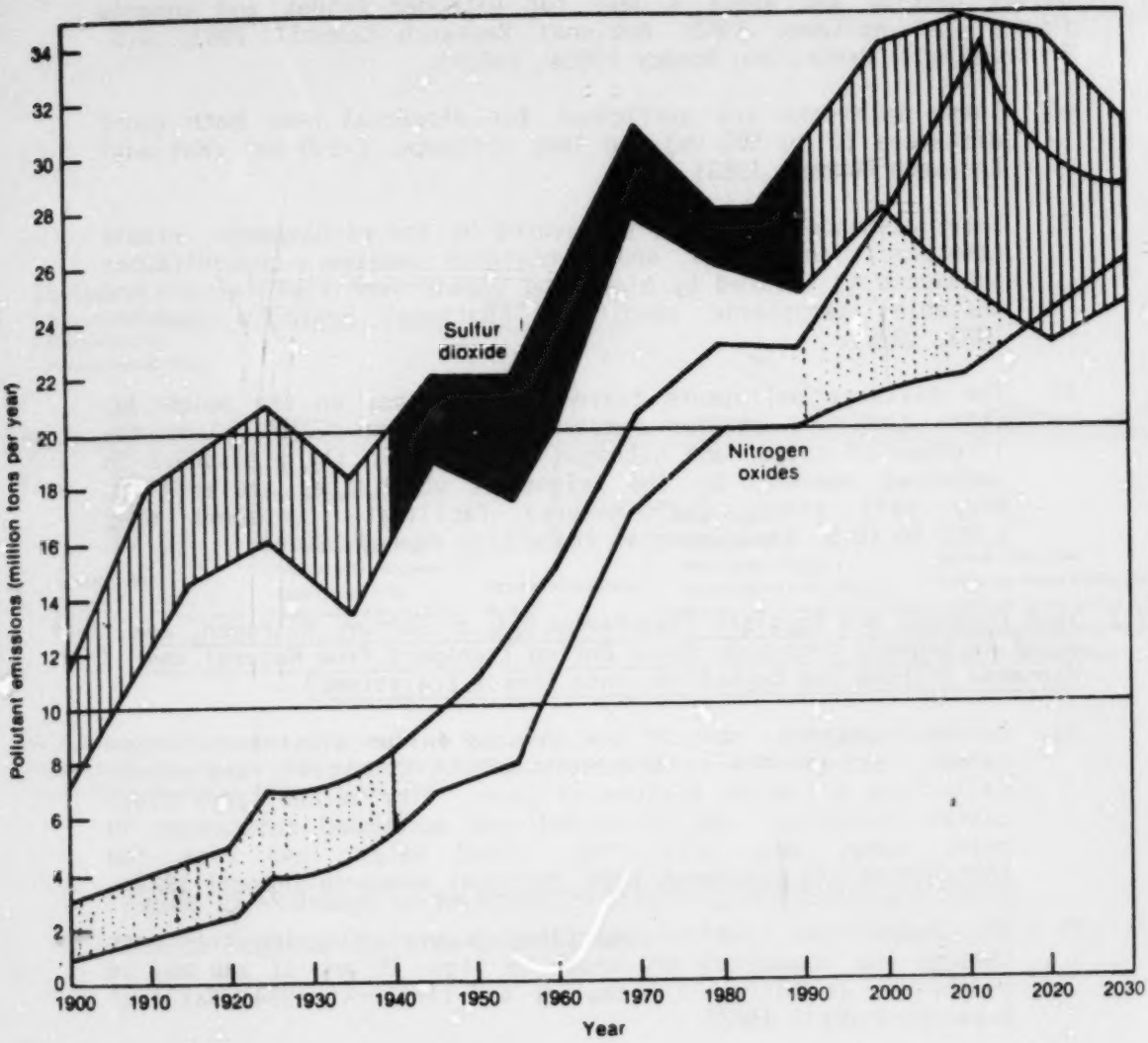


Figure 9. Sulfur dioxide and nitrogen oxides emissions trends—national totals, 1900–2030. The graph above displays estimates of historical emissions, and projections of future emissions of sulfur dioxide and nitrogen oxides. Pre-1940 estimates and post-1990 projections are subject to considerable uncertainty. Projections of future emissions incorporate a wide range of assumptions about future economic growth, energy mix, and retirement of existing facilities; they assume no change in current air pollution laws and regulations.

Source: U.S. Congress (1984).

sulfur dioxide and about 4 days for nitrogen oxides and ammonia (Husar and Holloway 1983; National Research Council 1983; U.S. Environmental Protection Agency 1982a, 1982b).

6) These intervals are sufficient for dispersal over both short distances (0 to 500 km) and long distances (>500 km) (National Research Council 1983).

7) Short-distance transport is favored by low wind speeds, stable atmospheric conditions, and mountainous terrain. Long-distance transport is favored by high wind speeds over flat terrain and unstable atmospheric conditions (National Research Council, 1983, 1986).

8) The distance pollutants travel also depends on the height at which they are emitted into the atmosphere. Generally, the lifetime of sulfur and nitrogen compounds in the atmosphere is increased somewhat by the height at which they are emitted. Very tall stacks (>250 meters) facilitate transport over 1,000 km (U.S. Environmental Protection Agency 1982b).

2.7 What Chemical and Physical Transformations of Sulfur, Nitrogen, and Volatile Organic Compounds Occur During Transport from Natural and Man-made Sources and Deposition into Forest Ecosystems?

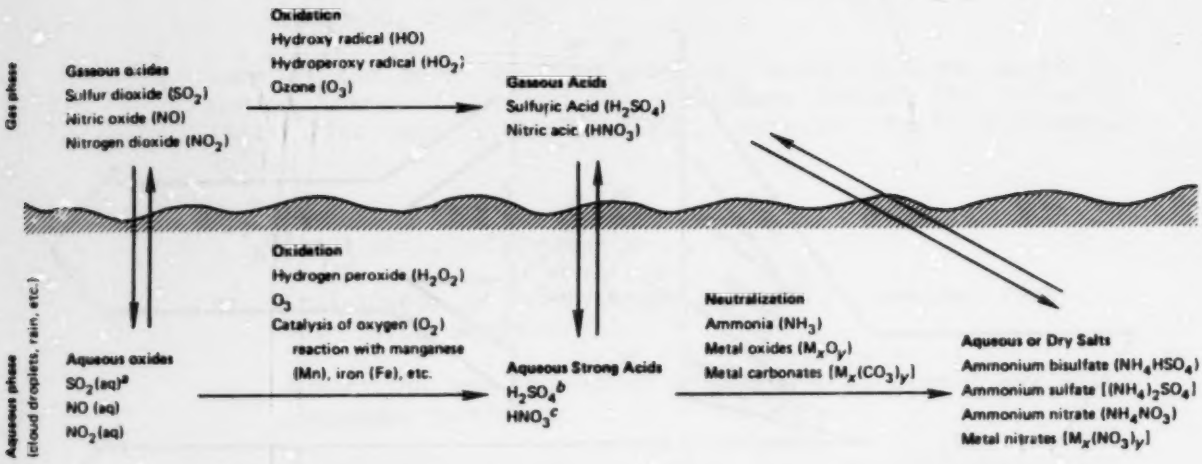
1) During transport, some of the gaseous sulfur dioxide, nitrogen oxides, and ammonia is transformed both chemically and physically into a complex mixture of gases, fine aerosol particles, coarse particles, and dissolved and suspended substances in rain, snow, dew, hail, fog, cloud water, and rime ice (Altshuller and Linthurst 1984; National Research Council 1983).

2) The predominant chemical reactions occurring during transport through the atmosphere are shown in figs. 10 and 11 and may be summarized as follows (Altshuller and Linthurst 1984; National Research Council 1983).

   a) Gaseous sulfur dioxide (SO_2) is further oxidized by airborne hydroxyl radicals ($OH^{\cdot}$) to sulfite ion (SO_3^-), which in turn reacts very rapidly with water or airborne particles, producing H_2SO_4 or sulfate particles such as ammonium sulfate [$(NH_4)_2SO_4$].

   b) Gaseous nitric oxide (NO) is quickly oxidized by airborne hydroxyl radicals ($OH^{\cdot}$) to gaseous NO_2. This reaction occurs so rapidly that almost all gaseous nitrogen oxides occur as NO_2.

   c) Gaseous NO_2 is further oxidized by airborne hydroxyl radicals ($OH^{\cdot}$) to form nitrate (NO_3^-), which combines with water or airborne particles to form HNO_3 or nitrate particles such as ammonium nitrate (NH_4NO_3).



[a] SO_2(aq) reacts to form hydrogen ions (H^+) and bisulfite ions (HSO_3^-).
[b] Aqueous H_2SO_4 dissociates into hydrogen ions, bisulfate ions (HSO_4^-), and sulfate ions ($SO_4^=$).
[c] Aqueous HNO_3 dissociates into H^+ and nitrate ions (NO_3^-).

Figure 10. Pathways for the formation of atmospheric sulfate and nitrate. Source: National Research Council (1983).

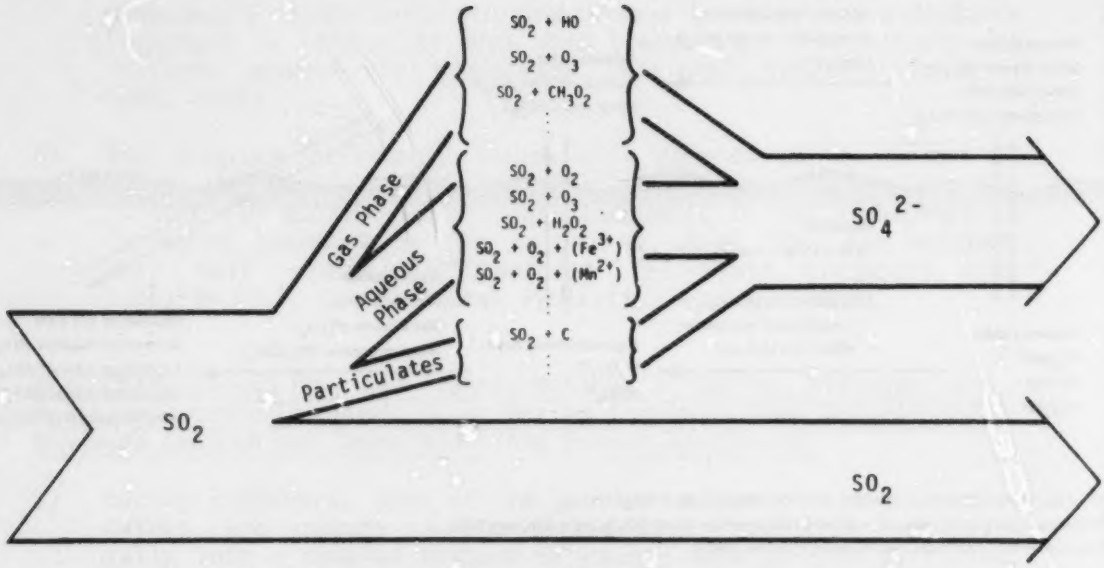


Figure 11. Sample processes in SO_2 oxidation. This figure illustrates the complexity of the atmospheric processes.

Source: Bennett and others (1985).

d) Gaseous nitrogen dioxide (NO_2) reacts with volatile hydrocarbons [volatile nonmethane organic compounds (VNMOC)] in the presence of sunlight to give a variety of highly reactive photochemical oxidants including ozone (O_3), hydrogen peroxide (H_2O_2), formaldehyde (H_2CO), peroxyacetyl nitrate (PAN) and peroxypropionyl nitrate (PPN).

e) Gaseous ammonia (NH_3) reacts with water to form ammonium ion (NH_4+), which may react with sulfate or nitrate to form $(NH_4)_2SO_4$ or NH_4NO_3.

3) Some of the more important physical transformations occurring during transport through the atmosphere include the following (Altshuller and Linthurst 1984; National Research Council 1983):

a) Formation of liquid and solid suspended particles by condensation of gases;

b) Coalescence of fine liquid and solid suspended particles into larger aerosols;

c) Adsorption and absorption of gases onto liquid and solid suspended particles;

d) Formation of clouds, as water vapor becomes supercooled;

e) Formation of raindrops, snowflakes, and hail;

f) Dissolution of gases in raindrops, snowflakes, and hail particles; and

g) Evaporation.

4) The end result of all the natural and man-made emissions, and the chemical and physical transformation processes during transport, is formation of a complex mixture of airborne chemicals that are present in the atmosphere as (Altshuller and Linthurst 1984; National Research Council 1983):

a) Gases;

b) Suspensions of fine aerosol particles (<2 microns in diameter) and larger aerosol particles (2-10 microns in diameter);

c) Suspensions of fly ash, soot, and other coarse particulate matter (10 microns to 1 millimeter or larger particles);

d) Dissolved and suspended substances in cloud water, fog, and precipitation including rain, snow, hail, fog, dew, and rime ice.

3. CHEMICAL EXPOSURE OF FORESTS IN EASTERN NORTH AMERICA

3.1 Introduction

Chapter 2 of this document describes the major sources, magnitude of emissions, principal chemical transformations, and the usual directions and distances over which sulfur- and nitrogen-derived compounds are dispersed through the atmosphere over eastern North America. Many meteorological, climatological, chemical, and biological factors influence the transfer of these compounds and their derived products from the atmosphere into forest ecosystems. These factors also cause large-scale spatial and temporal variation in concentration, residence time, and deposition of these substances across the continent and smaller-scale variations associated with differences in elevation.

The major purpose of the present chapter is to define and quantify the nature and magnitude of the chemical exposure of forests in various regions of eastern North America. The chemical form of the pollutant, concentration, duration, time of day and season of year are all important in determining exposure and response of forests.

3.2 What are the Most Important Airborne Sulfur- and Nitrogen-derived Pollutant Chemicals to which Forests are Exposed in Eastern North America?

1) The airborne sulfur- and nitrogen-derived chemicals presently known or suspected to cause growth-stimulating or growth-inhibiting effects on forests through foliage-mediated response mechanisms include the following (U.S. Environmental Protection Agency 1982a, 1982b, 1986):

   - Gaseous sulfur dioxide (SO_2);
   - Gaseous nitrogen oxides (NOx), mainly NO_2 but also NO;
   - Gaseous peroxyacetyl nitrate (PAN);
   - Gaseous peroxypropionly nitrate (PPN);
   - Gaseous nitric acid;
   - Gaseous ammonia (NH_3);
   - Gaseous ozone (O_3);
   - Gaseous hydrogen peroxide (H_2O_2);
   - Hydrogen peroxide dissolved in clud water precipitation;

2) The next four ionic materials occur mainly as fine aqueous aerosol particles and as dissovled and suspended materials in cloud water and precipitation. Their main effect on forest growth is through depostion on the soil.

- Sulfate ions ($SO_2^=$);
- Nitate ions (NO_3^-);
- Ammonium ions (NH_4^+);
- Hydrogen ions (H^+);

All of these last four ionic materials occur mainly in two forms: fine aqueous aerosol particles; and dissolved and suspended materials in cloud water and precipitation.

3) Gaseous peroxyacetyl nitrate (PAN); measurement methods for the sulfur- and nitrogen-derived pollutants are described in the Criteria Documents for nitrates, sulfates, and ozone (U.S. Environmental Protection Agency 1973, 1982b, 1986). The majority of air pollution monitoring devices in eastern North America have been placed in urban areas. The only monitors located specifically in forests were the ozone monitoring sites of the Environmental Protection Agency-Forest Service and National Park Service monitoring programs.

3.3 By What Physical Processes are Airborne Pollutant Chemicals Transferred from the Atmosphere into the Forests of Eastern North America?

The rate of transfer of airborne pollutant chemicals into forests is determined by a variety of chemical, physical, and meteorological features of the pollutants and their dispersal in the atmosphere of the forest, and by various physical, chemical, and biological features of plants, animals, and microorganisms in the ecosystems themselves. These several atmospheric and biological factors are important in assessing exposure and are discussed in the following sections.

1) Pollutant chemicals are dispersed in the atmosphere in four physical states (Hales 1984; National Research Council 1983):

- as gases;
- as fine aerosol particles (<2 microns in diameter) and coarse aerosol particles (2 to 10 microns in diameter). In the humid environment of eastern North America, most aerosols occur as tiny liquid particles, very few as solid particles;
- as fly ash and other coarse particulate matter (10-micron to 1-mm or larger particles); and
- as dissolved and suspended substances in cloud water, fog water, and precipitation.

2) These four types of substances are transferred from the atmosphere into forest ecosystems where they are deposited onto two types of surfaces (Hales 1984; Hicks 1984): solid plant, animal, microbial, soil, and stone surfaces; and liquid water surfaces within forest ecosystems.

The following physical mechanisms of transfer and deposition are involved:

- Gases are transferred to solid surfaces by adsorption, absorption, condensation, and diffusion processes. They also are transferred to dew and other liquid water films, water droplets, and pools of liquid water by condensation, dissolution, and diffusion processes.

- Fine and coarse aerosol particles are transferred mainly by physical impaction processes onto both the solid and liquid surfaces in forests.

- Fly ash and other coarse particulate matter are transferred into forests by gravitational settling.

- Dissolved and suspended substances in precipitation are transferred into forests by physical impaction in the case of cloud water, fog water, and rime ice; and by gravitational settling in the case of raindrops, snowflakes, and hail and sleet particles.

3) Gases, fine and coarse aerosols (including fog and cloud water droplets), and coarse particulate matter are transferred into forests by dry deposition processes. These dry deposition processes occur under all meteorological conditions, at all times of the day and night, and in all seasons of the year (Hicks 1984).

4) The rate of transfer of gases and aerosols from the atmosphere into forests (deposition velocity) is a complex function of the following (Fowler 1980):

- concentration of gases and aerosols in the atmosphere which in turn depend on:

  - proximity to and strength of major pollution sources;
  - duration and frequency of air stagnation events (see fig. 12);
  - time since the most recent precipitation event; and
  - thoroughness of atmospheric cleansing achieved during that event;

- speed and turbulence of wind;

- size and aerodynamic properties of aerosol particles;

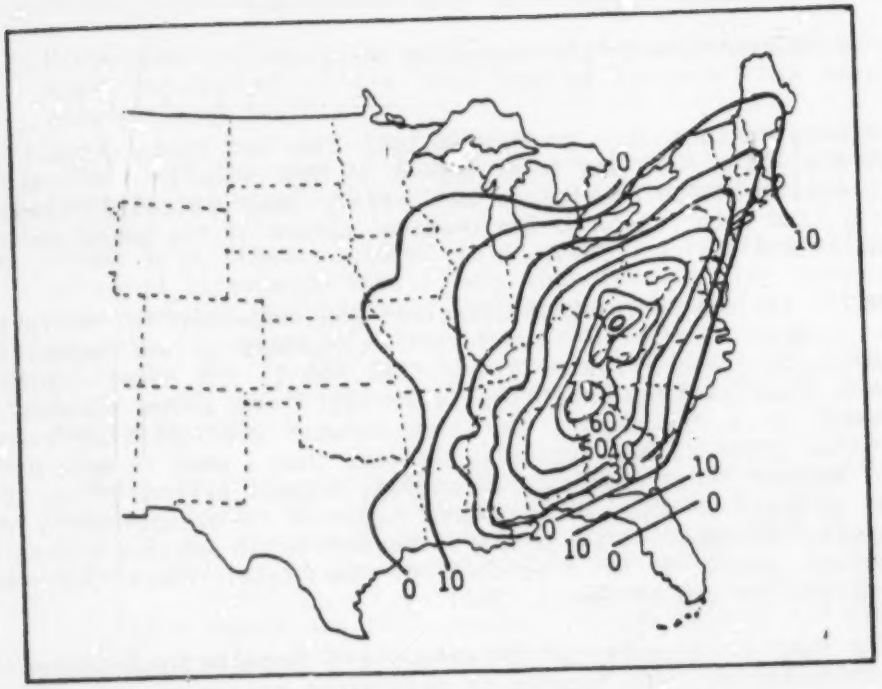


Figure 12. Incidence of air stagnation advisories issued over a 10-year period. Adapted from Lyons (1975).

Source: Bennett and others (1985).

- chemical and physical properties of gases and aerosols;
- chemical and physical properties of the surfaces to which the gases and aerosols are transferred;
- temperature and relative humidity of the free atmosphere, and of the atmosphere near the surface to which the gases and aerosols are being transferred; and
- temperature of the receiving surface.

Since these many factors vary greatly in both time and space (especially in complex terrain), the rate and total amount of dry deposition of gases and aerosols in eastern North America varies greatly geographically, temporally, altitudinally, and with regard to the chemical nature of the gases and aerosols involved (Hales 1984; Hicks 1984).

5) Major cations and anions dissolved and suspended in precipitation are transferred into forest ecosystems by wet deposition processes (i.e., during rain, snow, hail, and sleet storms). Wet deposition episodes typically last from a few minutes or hours to a few days. The time between precipitation events varies greatly, but typically is less than 1 week in most parts of eastern North America. Of course, drought periods of up to 7 or 8 weeks or even longer have occurred in exceptionally dry years. Wet deposition events in eastern North America generally occupy about 4 to 8 percent of the total hours per year (Altshuller and Linthurst 1984).

6) The rate of transfer of dissolved and suspended substances in precipitation into forests is determined mainly by the concentration of these substances in the atmosphere and the amount of precipitation that occurs per unit time. These substances are removed from the free atmosphere by cloud nucleation processes (rain-out), and by impaction processes as raindrops, snowflakes, and hail and sleet particles fall through the atmosphere below clouds (wash-out) (Altshuller and Linthurst 1984).

3.4 What Physical and Meteorological Features of the Terrain on which Forests Grow and the Physical Form of Forest Canopies Influence the Amounts of Airborne Pollutant Chemicals that are Delivered into Forest Ecosystems in Various Parts of Eastern North America?

1) The magnitude of pollutant load received by a forest is generally an inverse function of the distance from major sources of pollution (Altshuller and Linthurst 1984; Fowler 1980).

2) The magnitude of pollutant load received by a forest on sloping terrain is generally greater on slopes facing predominant winds than on slopes facing in other directions, especially those facing away from predominant winds (Altshuller and Linthurst 1984).

3) Forests with very uneven canopy heights (such as the mixed conifer-hardwood forests of New England and southeastern Canada) generally receive heavier pollutant loads than forests that have very even canopy heights (such as the beech-birch-maple forests of New England and southeastern Canada or the naturally regenerated pine-oak-hickory forests or planted stands of southern pine in the Piedmont and Coastal Plain regions of the Southern United States) because they have a larger number of impaction surfaces.

4) Mixed-species forests generally receive heavier pollutant loads than forests of single tree species because they have very uneven canopy heights.

5) Trees near the edge of forest stands generally receive heavier loads of pollutants than trees within forest stands.

6) Trees with stomata closed (high stomatal resistance) take up airborne gases much more slowly than forests with stomata open (low stomatal resistance). Typically, 4- to 20-fold differences in rate of uptake of gases are observed, for example, during night and day in the case of tree species that close stomata at night, and during droughts compared to wet periods (Mudd and Kozlowski 1975).

7) Forests on high mountains (> 1000 meters) are exposed to much heavier loads of airborne pollutant chemicals than similar forests at low elevation (Lovett and others 1982; Wolff and others 1987). This is caused by the following:

- Wind speeds are much greater at high than at low elevation (typically 2 to 10 times greater);
- Amounts of precipitation are generally much greater at high than at low elevation (typically 2 to 3 times greater);
- Fog and clouds occur much more frequently at high than at low elevation (typically 2 to 10 times greater);
- Fog and cloud water significantly augment the total amount of water delivered to forest canopies through precipitation processes (typically by 20 to 200 percent);
- Concentrations of dissolved and suspended substances in fog and cloud water are typically 5 to 10 times greater than those in precipitation;
- Concentrations of ozone and other photochemical oxidants are often greater at high than at low elevation (typically about 10 to 50 percent greater); and
- Diurnal periodicity in concentration of photochemical oxidants, especially ozone, is much less at high than at low elevations.

3.5 In What Concentrations and During What Times of Day and Seasons of the Year are Sulfur- and Nitrogen-derived Compounds Deposited in Forests in Various Regions of Eastern North America?

Proximity to pollutant sources is important in determining the chemical exposure of forests in various regions of North America. The general contrast in concentrations of sulfur dioxide, nitrogen oxides, and ozone in urban, suburban, rural, and remote regions of the northeastern States is shown in fig. 13 (Barchet 1987).

In our efforts to characterize the chemical exposure of forests in various parts of eastern North America, we found it useful to recognize five general types of forests based on proximity to sources:

1) Urban forests such as the 30,000 hectares of forest land within the city limits of Atlanta, GA. Although urban forests probably constitute a very small percentage of the total area of eastern North America, urban areas are home to about 30 percent of the human population of eastern North America and thus are of great interest to many people in the United States and Canada;

2) Rural forests near major point sources of pollution (for example, within about 30 km of a major power plant or metal smelter). We estimate that these areas make up about 5 percent of the total forest area of the eastern United States and southeastern Canada;

3) Near-urban forests within about 50 km of major sources of pollutant areas. We estimate that these areas include about 37 percent of the total forest area of the eastern United States and southeastern Canada;

4) Rural forests more than 50 km distant from major urban areas or from major point sources of pollution. We estimate that these areas make up about 25 to 30 percent of the total forest area of the eastern United States and southeastern Canada; and

5) High-elevation forests such as those on Mount Mitchell, NC, and Whiteface Mountain in New York (see fig. 14). These areas are characterized by frequent exposure to clouds and concentrations of ozone that show little or no diurnal variability. We estimate that these areas make up about 2 percent of the total forest area of the eastern United States and southeastern Canada.

In making these distinctions we have followed the lead of the personnel of the National Vegetation Survey of the Forest Response Program who have been developing the so-called Southern Forest Atlas/Geographic Information System, in particular the "Chemical Atlas". They have used aerial photographs in assessing the proximity of the U.S. Environmental Protection Agency (EPA), Electric Power Research Institute (EPRI), and Tennessee Valley Authority (TVA) monitoring sites to forests and to major point and area sources of airborne pollutant chemicals. Although we have been unable to do so thoroughly in the

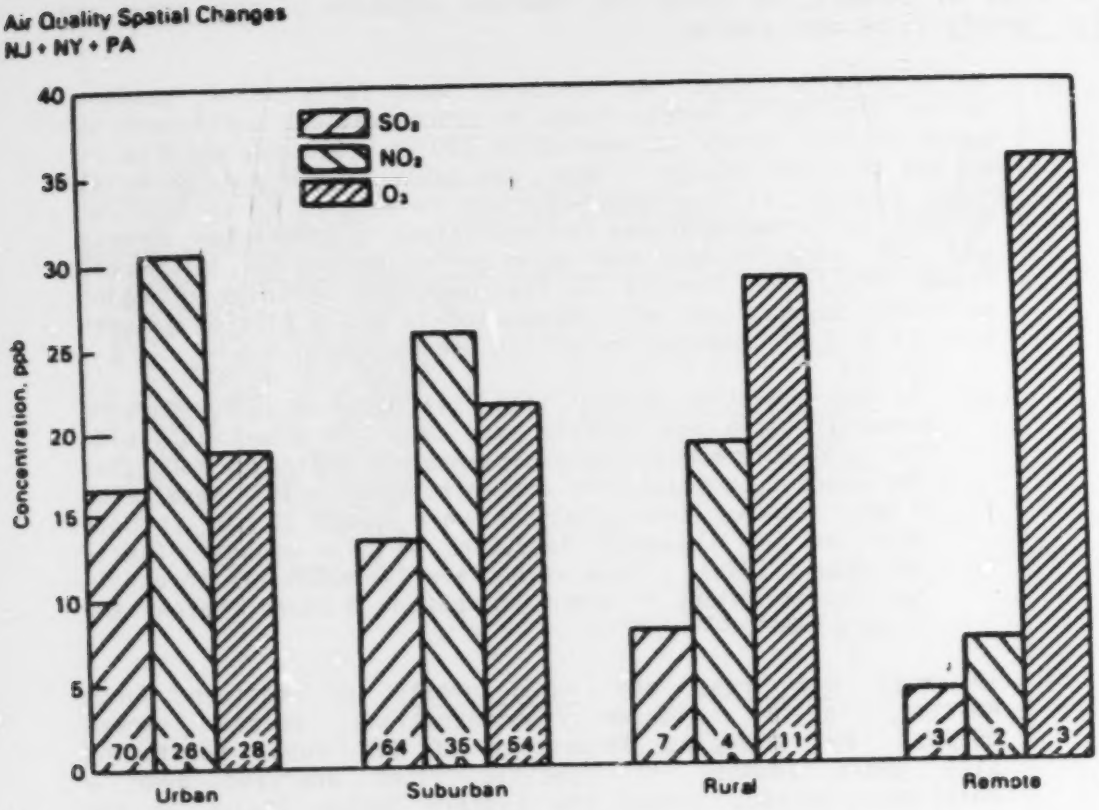


Figure 13. Mean annual air quality for SO_2, NO_2, and O_3 by station class in the three-state region of New Jersey, New York, and Pennsylvania for the period 1976 to 1983. The number on the vertical bar indicates the number of stations in that class. 1 ppm SO_2 = 2600 $\mu g/m^3$; 1 ppb NO_2 = 1900 μ/m^3.

Source: Barchet (1987).

time available, we believe aerial-photograph interpretation of the amounts and types of forests near monitoring sites offers a useful means by which to increase further our understanding of the chemical exposure of forests of the five types listed above.

In drawing certain general inferences about the chemical exposure of these five types of forests, we found the chemical exposure of near urban and rural forests to be very similar.

1) Sulfur dioxide (SO_2): As discussed more fully below, gaseous sulfur dioxide is rarely found in concentrations sufficient to cause visible injury to vegetation (50 to 5,000 ppb for 8 hours or 500 to 3,000 ppb for 1 hour, see table 3) in remote forests, rural forests, or near-urban forests in eastern North America. It does occur in injurious concentrations in some urban forests and in rural forests near major pollution sources, however. These effects on forests in the immediate vicinity of major pollution sources are well documented in the published literature (U.S. Environmental Protection Agency 1982a).

   a) In high-elevation forests (fig. 14), gaseous sulfur dioxide concentrations are usually very near the detection limits for available monitoring equipment. At Whiteface Mountain, for example, average 1-hr concentrations in 1982 were below 1 ppb; maximum concentrations were about 20 ppb at both high and low elevation locations on this mountain (Kelly and others 1984). These values are far below the threshold for foliar injury -- about 50 ppb for 8 hours (Lefohn and Tingey 1984).

   b) In most near-urban and rural forests of eastern North America, sulfur dioxide concentrations seldom exceed 50 ppb. For example, 39 percent of 922 rural monitoring site years (number of specific sites analyzed over a multi-year period) across the Eastern United States in the Environmental Protection Agency SAROAD, EPRI, and TVA monitoring networks for 1978-81 showed no 1-hour average sulfur dioxide concentrations greater than or equal to 50 ppb (Lefohn and Tingey 1984). Similarly, 90 percent of all 1-hour average values for sulfur dioxide at 20 sites classified as rural or remote in the SAROAD data base for 1979-83 in the States of Arkansas, Louisiana, Mississippi, Georgia, and Florida were less than 10 ppb. The highest 1-hour average concentration observed at any of these 20 sites was 822 ppb. Only two sites showed 1-hour average values greater than 500 ppb. Eleven sites showed no values greater than 200 ppb even for a single hour in 1979-83 (Lefohn and others 1987). This is true in part because sulfur dioxide is rapidly transformed to sulfate aerosol during transport from major sources (Husar and Holloway 1983).

Figure 14. Regions of the United States with high-elevation forests (boreal, sub-alpine, and montane; mixed boreal and deciduous; mixed boreal, lake, and deciduous).

Source: Bennet and others (1985).

c) Some urban forests and rural forests near major pollution sources in eastern North America are exposed to phytotoxic concentrations of gaseous sulfur dioxide. Maximum concentrations of 500 to 1,000 ppb or even higher have been reported near some industrial sources for several days at a time during the 1970's and early 1980's (U.S. Environmental Protection Agency 1982a).

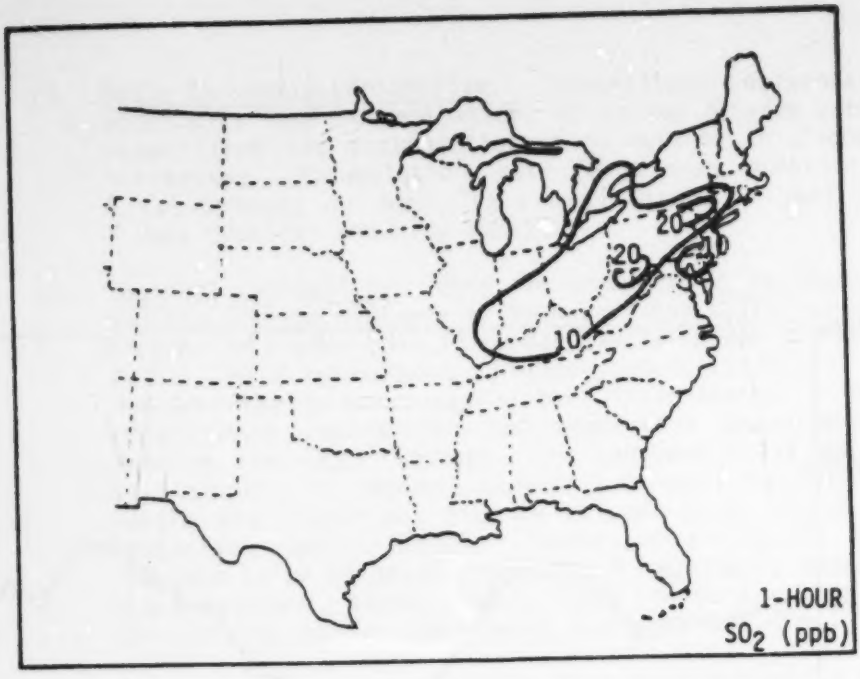
In Electric Power Research Institute's Sulfate Regional Experiment in 1977-78, the highest annual average concentrations of sulfur dioxide observed in rural areas was 20 to 30 ppb (see fig. 15). These relatively high values were observed in the Ohio River Valley and in western Pennsylvania. The highest monthly 1-hour average values of about 30 ppb occurred during January and February 1978. The highest concentrations in near-urban areas were found in the vicinity of Richmond, VA, Philadelphia, PA, New York, NY, and Boston, MA (Mueller and others 1980).

d) Regional variability. The region with the highest annual average sulfur dioxide concentrations at rural sites in eastern North America is in the Ohio River Valley where these values approach 10 ppb. Average concentrations decrease with increasing distance from this region, reaching approximately 2 ppb in New England and less than 1 ppb in the Southern United States (Mueller and Hidy 1983).

In the Environmental Protection Agency SAROAD data base for 1980-85, the ranges of sulfur dioxide concentrations at representative low-elevation rural and remote sites in the Northeastern and Southeastern United States were as follows:

	Northeast	Southeast
Number of sites	11	16
24-hr arithmetic mean	7 - 13 ppb	7 - 8 ppb
24-hr maximum concentration	14 - 57 ppb	3 - 52 ppb
1-hr maximum concentration	29 - 210 ppb	5 - 200 ppb

e) Diurnal variability. The highest 1-hour concentrations of sulfur dioxide in near-urban and rural forests usually occur at night and during the early-morning hours if emissions are predominantly from residential and industrial space-heating sources with low stacks (fig. 16) (U.S. Environmental Protection Agency 1982a). When emissions are from tall stacks, however, highest concentrations usually occur in the morning just after sunrise with a broad diurnal maximum between 4 p.m. and 6 a.m. (U.S. Environmental Protection Agency 1982a).



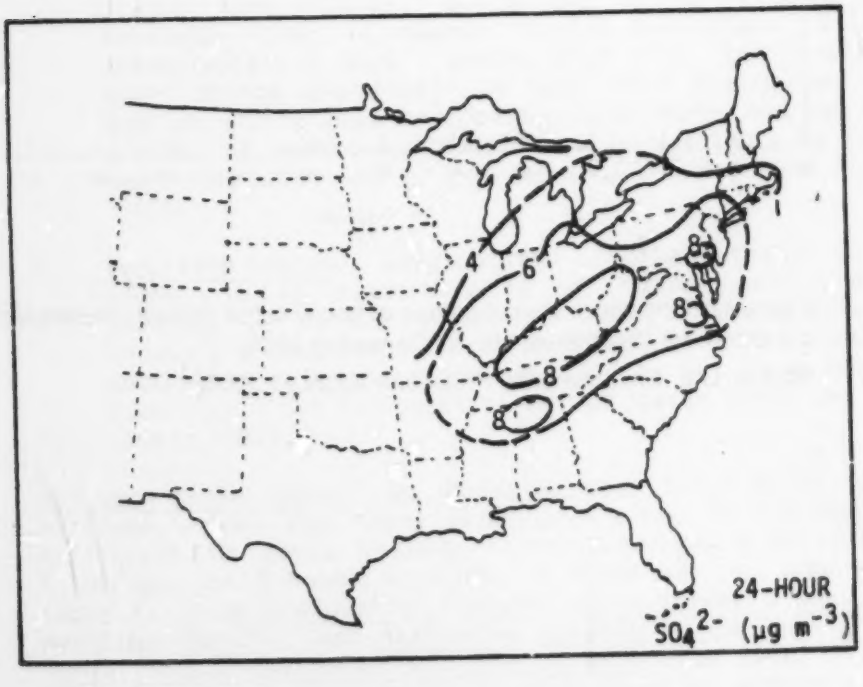


Figure 15. Sulfur dioxide (arithmetic mean) and sulfate (geometric mean) concentrations. Data obtained during 5 months between August 1977 and July 1978. Note: 10 ppb SO_2 is approximately 28 $\mu g/m^3$ SO_4.

Source: Bennett and others (1985).

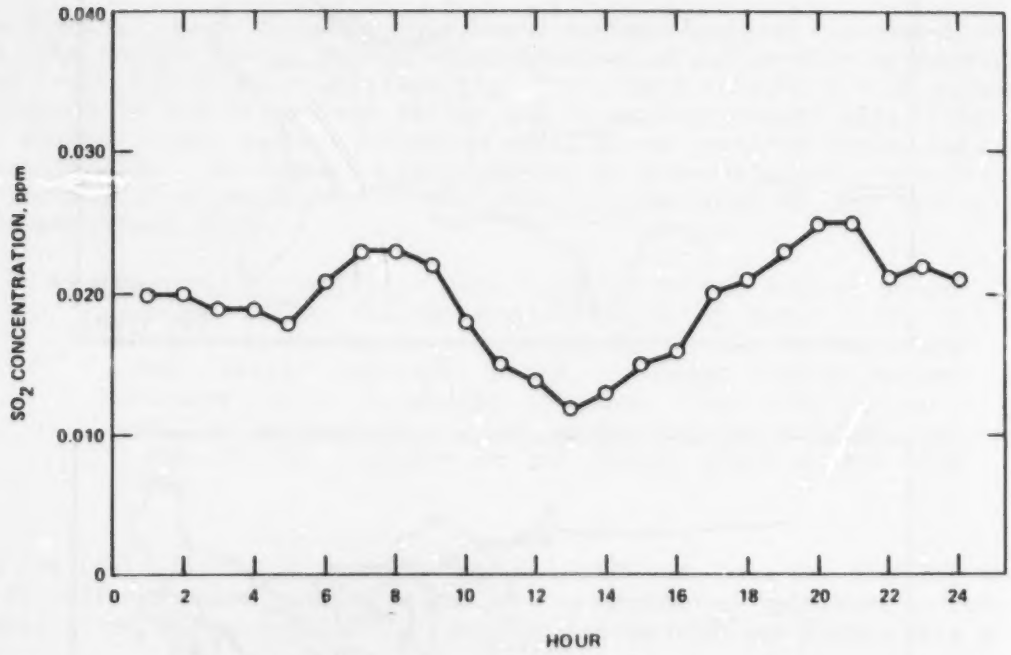


Figure 16. Composite diurnal pattern of hourly sulfur dioxide concentrations are shown for Watertown, MA, for December 1978.

Source: U.S. Environmental Protection Agency (1982a).

f) Daily to weekly variability. Several-day episodes with atypically high concentrations of sulfur dioxide sometimes result from the accumulation of pollutants in slow-moving air masses. Accumulations over large areas sometimes last 3 to 10 days, or when the area involved is small, 1 to 7 days (Mueller and Hidy 1983).

g) Seasonal variability. Both the atmospheric residence time and the average distance of transport for sulfur dioxide is greater in winter than in summer months (Husar and Holloway 1983). As a consequence, highest concentrations of sulfur dioxide usually occur during the winter months in northern areas where residential and commercial space and water heating are major sources. In southern areas where air conditioning is common, however, a small secondary peak during the summer may also be evident (U.S. Environmental Protection Agency 1982a). Summer concentrations are are presumed to be of greater biological importance than winter concentrations, since forest trees are more active during the growing season (Kozlowski and Constantinidou 1986a).

h) Major sources. Approximately 70 percent of sulfur dioxide emissions in the Eastern United States are from power plants, many with tall stacks and located in rural areas. Residence time in summer in the midwestern States is approximately 2 days. During that time, the plumes from power plants are likely to have been transported up to 1000 km and to have broadened considerably, and the sulfur dioxide to have been transformed into sulfate or to have been removed by wet or dry deposition (Husar and Holloway 1983).

i) Long-term trends. Judging from long-term trends in emissions, it is reasonable to infer that sulfur dioxide loadings of forest ecosystems have remained relatively constant or decreased in the Northeastern United States and southeastern Canada since the 1920's, but have increased steadily in the Southeastern United States since the 1940's (Husar 1986).

2) Nitrogen oxides (NOx): As discussed more fully below, gaseous nitrogen oxides are found rarely, if ever, in concentrations sufficient to cause visible injury to vegetation (2,000 to 5,000 ppb for 8 hours or 4,000 to 8,000 ppb for 1 hour, (see table 3) in high-elevation forests, rural forests, coastal and maritime forests, and near-urban forests or even in urban or rural forests near major pollution sources. In fact, injury to trees or other forest vegetation by exposure to gaseous nitrogen oxides occurs so rarely in eastern North America that there are no reports of injury in the published literature (National Research Council 1977b; U.S. Environmental Protection Agency 1982b) (see also chapter 4).

a) In high-elevation forests, concentrations of gaseous nitrogen oxides usually are below the detection limits for available monitoring equipment. At Whiteface Mountain, for example, average 1-hour concentrations in 1982 were below 1 ppb, and maximum concentrations were about 3 ppb (Kelly and others 1984). At a series of remote sites in the Eastern United States in 1978-81, 1-hour NOx concentrations typically exceeded 50 ppb fewer than 10 times per year (Lefohn and Tingey 1984). These values are far below the threshold for visible injury -- about 2000 ppb for 8 hours (U.S. Environmental Protection Agency 1982b).

b) In most near-urban and rural forests of eastern North America, concentrations of nitrogen oxide seldom exceed 10 ppb (U.S. Environmental Protection Agency 1982b). This is true in part because nitrogen oxides are often consumed in the photochemical formation of ozone (see sec. 2.6). Annual average 1-hour NOx concentrations for 9 rural EPRI sites in the Eastern United States were 7 ppb from 1978 to 1982. The only EPRI site where significant nitrogen oxides were recorded at a rural site was near Philadelphia, PA (Mueller and Hidy 1983). There, the maximum nitrogen oxides concentration was 150 ppb.

c) Significant nitrogen oxides concentrations are usually confined to urban areas and rural areas near major pollution sources. For example, in 1976-1980, peak concentrations of 270-ppb nitrogen oxides were reported at St. Louis, MO; New York, NY; Springfield, IL; Cincinnati, OH; and Saginaw and Southfield, MI (U.S. Environmental Protection Agency 1982b). Similarly, during the years 1976-1980, peak NOx concentrations equaling or exceeding 400 ppb were measured at rural industrial areas near Ashland, KY, and Port Huron, MI. Annual average concentrations in urban areas of eastern North America are typically about 70 ppb (U.S. Environmental Protection Agency 1982b).

Urban nitrogen oxide concentrations between 6:00 and 9:00 a.m. are very important in the formation of ozone and other photochemical oxidants. Average nitrogen oxides concentrations during these hours ranged from about 50 to 150 ppb in nine urban areas (U.S. Environmental Protection Agency 1982b).

d) Regional variability. The regions with the highest annual average nitrogen oxide concentrations at rural and remote sites in eastern North America are in the Ohio River Valley and in the southern Piedmont regions of the southeastern States; here these values approach 12 ppb. Average concentrations decrease with increasing distance from these regions, reaching approximately 1 ppb in remote areas of New England and elsewhere in the Southern United States (Mueller and others 1980).

In the Environmental Protection Agency SAROAD data base for 1980-85, the ranges of nitrogen oxide concentrations at representative low-elevation rural sites in the Northeastern and Southeastern United States were as follows:

	Northeast	Southeast
Number of sites	8	16
24-hr arithmetic mean	10 - 20 ppb	3 - 17 ppb
24-hr maximum concentration	27 - 62 ppb	12 - 75 ppb
1-hr maximum concentration	27 - 338 ppb	21 - 100 ppb

e) Diurnal variability. No typical diurnal patterns of nitrogen oxide concentrations exist. Although a recurrent diurnal pattern is discernible in some areas of the United States, in many other areas peak concentrations can occur at almost any time of day or night. Concentrations of nitrogen oxides in urban areas, like those of volatile hydrocarbons, tend to increase rapidly in the morning as a result of NO emissions and photochemical conversion to NO_2 when atmospheric dispersion is limited and automobile traffic is dense. Photochemical generation of nitrous oxides is followed by a concomitant rapid increase in ozone concentration, which depresses the nitrous oxide concentrations until later afternoon hours, when decreasing radiant energy and increasing NO emissions overwhelm the ozone-generating mechanisms. High NO_2 concentrations then occur after photochemical activity has ceased (fig. 17). Short-term peaks of NO_2 can occur at night and are not associated with traffic emissions or photochemical oxidation (U.S. Environmental Protection Agency 1982b).

f) Daily to weekly variability. Several-day episodes with atypically high concentrations of nitrogen oxide sometimes result from the accumulation of pollutants in slow-moving air masses. Accumulations over large areas sometimes last 3 to 10 days, or when the area involved is small, 1 to 7 days (Mueller and Hidy 1983).

g) Seasonal variability. Just as there is no typical diurnal pattern for peak nitrogen dioxide concentrations, there is also no typical seasonal pattern. Peak monthly averages occur at different times of the year in different locations. In remote areas, nitrogen oxide and nitrous oxide seasonal peaks occurred any time between October and April (U.S. Environmental Protection Agency 1982b). Power plants emit about 15 percent more nitrogen oxides in summer than in winter. Also, seasonal variation in automobile travel leads to seasonal variation in emissions of nitrogen oxides -- 18 percent more vehicle miles are traveled in summer than in winter (U.S. Environmental Protection Agency 1986).

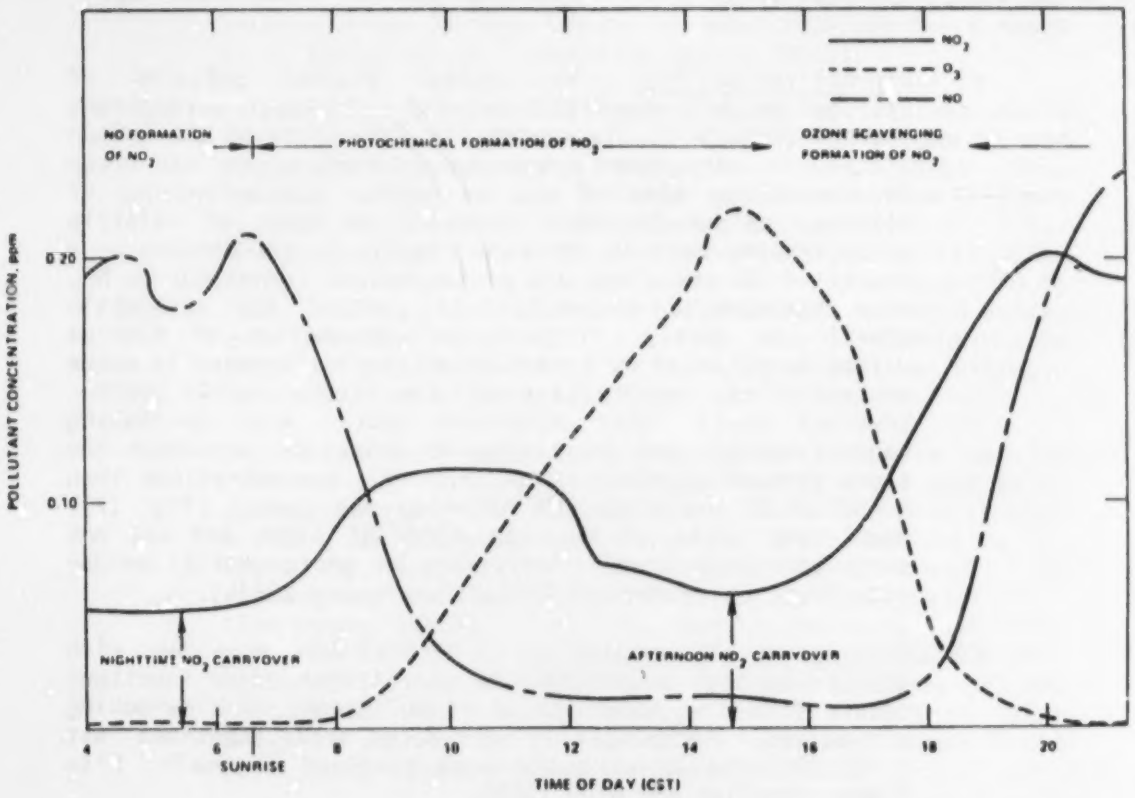


Figure 17. Pollutant concentrations in Central City, St. Louis, October 1, 1976, average of RAMS sites 101, 102, 106, and 107. Illustration of photochemical and ozone scavenging formation of NO_2.

Source: U.S. Environmental Protection Agency (1982b).

h) Major sources. Approximately 40 percent of nitrogen oxide emissions are from transportation vehicles, about 40 percent from power plants, and the remaining 20 percent from other industrial, commercial, and domestic sources (U.S. Environmental Protection Agency 1982b).

i) Long-term trends. Judging from long-term trends in emissions, it is reasonable to infer that nitrogen oxide loadings in forest ecosystems have increased markedly in most parts of eastern North America (Husar 1986) and will probably increase still further well into the 21st century (U.S. Congress 1984).

3) Ozone (O_3): As discussed more fully below, gaseous ozone is found in concentrations sufficient to cause visible injury to vegetation in many parts of eastern North America. For example, 20 to 40 ppb for 8 hours or 90 to 150 ppb for 2 hours (see table 3) have been reported in many urban forests, rural forests near major point sources, near-urban forests, rural forests, coastal and maritime forests, and in high-elevation forests. Ozone-induced visible injuries on forest trees in various parts of the continent are well documented in the published literature (U.S. Environmental Protection Agency 1986).

a) In high-elevation forests, average 7-hour summertime concentrations of ozone are only somewhat lower than those near major urban and rural pollution sources in the southern United States--for example, 30 to 32 ppb at monitoring sites in the Croatan National Forest in North Carolina and DeRidder, LA, compared to 37 to 57 ppb at 26 other sites listed in table 7 (Evans and others 1983; Pinkerton and Lefohn 1986). At some other remote sites, particularly at high elevations, however, 1-hour peak concentrations of gaseous ozone often are as high as those in some urban and near-urban forests. In the Shenandoah National Park, for example, peak summertime ozone concentrations in excess of 100 ppb were reported in the early 1960's (Berry 1964). Similarly high 1-hour peak concentrations have also been observed in most years between 1975 and 1984 at Whiteface Mountain (Lefohn and Mohnen 1986; Mohnen 1987). Furthermore, ozone shows little diurnal periodicity at high elevation sites; thus, high-elevation forests are exposed to high concentrations of ozone for many more consecutive hours than forests in low elevation locations. Seven-hour average ozone concentrations at high-elevation sites are frequently about double the concentrations of low elevations in the same geographical areas (Lefohn and Jones, 1986; Lefohn and Mohnen 1986). These values are substantially greater than the threshold for visible injury by ozone--20 to 40 ppb for 8 hours (Lefohn and Tingey 1984; U.S. Environmental Protection Agency 1986).

Table 7. Ozone concentrations in 1983 at rural sites located more than 50 km from a city with a population over 50,000

Monitoring Site	7-hr mean 0900-1559 April-October	Cumulative 90	Frequency 95	Percentiles[a] 99	Peak	Number of hourly average concentrations equal to one or greater than 0.07 ppm	0.09 ppm	0.12 ppm	0.14 ppm
VIRGINIA									
Fauquier Co.	0.055	0.068	0.077	0.097	0.129	351	166	27	7
Marion	0.055	0.070	0.070	0.090	0.110	490	172	7	0
Loves Mill	0.048	0.059	0.067	0.082	0.104	285	105	3	0
Browntown	0.054	0.075	0.080	0.090	0.110	965	427	16	0
Waynesboro	0.056	0.065	0.075	0.095	0.120	502	259	39	2
Skyland	0.055	0.070	0.075	0.090	0.110	719	222	16	0
NORTH CAROLINA									
Lenoir	0.051	0.065	0.074	0.087	0.118	368	157	12	0
Alexander Co.	0.053	0.066	0.075	0.090	0.113	314	129	6	0
Edgecombe Co.	0.057	0.073	0.083	0.098	0.123	475	251	29	1
Farmville	0.049	0.062	0.070	0.087	0.108	225	91	5	0
Martin Co.	0.050	0.064	0.072	0.087	0.102	169	74	2	0
Johnston Co.	0.053	0.066	0.076	0.088	0.105	211	85	4	0
Montgomery Co.	0.051	0.069	0.075	0.090	0.118	424	161	14	0
Croatan NF	0.038	0.045	0.055	0.070	0.085	93	11	0	0
SOUTH CAROLINA									
Chester Co.	0.058	0.066	0.077	0.096	0.142	655	311	52	6
TENNESSEE									
Giles Co.	0.051	0.059	0.069	0.083	0.107	341	106	7	0
KENTUCKY									
Calloway Co.	0.051	0.069	0.075	0.088	0.143	379	115	14	6
Land between Lakes	0.052	0.063	0.072	0.084	0.129	486	192	9	1
Fulton Co.	0.051	0.067	0.074	0.087	0.105	311	117	5	0
Russellville	0.041	0.058	0.065	0.080	0.114	124	47	9	0
Paradise	0.051	0.062	0.073	0.092	0.129	441	185	31	4
Metcalf Co.	0.051	0.066	0.077	0.090	0.109	245	127	10	0
Pulaski Co.	0.041	0.060	0.068	0.085	0.109	183	78	2	0
Clay Co.	0.040	0.059	0.068	0.083	0.107	179	64	4	0
Prestonburg	0.037	0.058	0.068	0.081	0.106	189	50	1	0
LOUISIANA									
DeRidder	0.032	0.045	0.052	0.068	0.116	20	4	2	0
Galliano	0.038	0.052	0.063	0.083	0.114	99	44	4	0
Morgan City	0.036	0.049	0.062	0.083	0.101	101	46	1	0

Source: Pinkerton and Lefohn (1986).

b) In many rural forests more than 50 km from a city with a population over 50,000 in the Southeastern United States, 7-hour average ozone concentrations in the summer months of 1983 ranged from 50 to 70 ppb with 1-hour peak concentrations sometimes exceeding 100 or even 120 ppb (see table 7) (Pinkerton and Lefohn 1986).

c) Although the 7-hour average concentrations in many urban forests, near-urban forests, and some rural forests near major point sources (see table 8) were about the same as those reported for rural forests (see table 7), this is not true of the highest 1-hr peak concentrations. For example, the data of table 8 show that 7-hour average summertime ozone concentrations for a selection of southeastern urban areas ranged from about 40 to 60 ppb--about the same as the 50 to 70 ppb shown for southeastern rural forests in table 8. But these same urban areas showed 1-hour peak concentrations in 1983 between 150 and 200 ppb -- substantially higher than the 102 to 143 ppb shown for rural forests in table 8. The data in table 8 also show that the six monitoring sites in Washington, DC, had ozone concentrations that exceeded 100 ppb for as long as 42 to 250 hours. Similarly, the two sites in Atlanta, GA, and Norfolk, VA, and the three sites in Louisville, KY, and Charlotte, NC, had ozone concentrations that exceeded 100 ppb for as long as 122 to 134 hours, 74 to 97 hours, 47 to 167 hours, and 26 to 133 hours, respectively (Pinkerton and Lefohn 1986). The 1980-85 Environmental Protection Agency SAROAD data for rural sites in the northeastern and midwestern States showed average and maximum concentrations that were similar to those measured in the southeastern States. Unfortunately, however, the number of sites in the southern States was much smaller than those in the northeastern and midwestern States (Evans and others 1983).

d) Regional variability. The highest 1-hour peak concentrations of ozone in the Eastern United States occur in New England, the mid-Atlantic States, and the Gulf Coast. From 1981 to 1983, peak concentrations of 170 ppb, 180 ppb, 250 ppb, and 280 ppb were reported for Raleigh, NC; Boston, MA; Newark, NJ; and Houston, TX, respectively (U.S. Environmental Protection Agency 1985).

The high concentrations of ozone that accumulate in urban, near-urban, rural, and even high-elevation areas are mainly associated with significant emissions of volatile organic compounds and nitrogen oxide precursors. Secondary pollutants such as ozone and other photochemical oxidants often are not formed until approximately 1 day after emission of their precursors. These high concentrations of ozone can be transported over distances of 100 to 300 km, during which time such air masses can pick up additional ozone as they travel over other urban areas. For example, transport of high concentrations

Table 8. Ozone concentrations in metropolitan areas in 1983

Metropolitan Area	1980 Population Millions	Number of Monitoring Areas	April - October 9:00 a.m. - 4:00 p.m. 7-hr mean	Number of Hourly Average Concentrations equal to or greater than ppm				Maximum Concentration ppm
				0.07	0.10	0.12	0.14	
Washington, DC	3.06	6	0.047 - 0.060	379 - 732	42 - 250	9 - 83	0 - 20	0.130 - 0.195
Houston	2.91	11	0.047 - 0.060	276 - 850	50 - 199	46 - 128	25 - 56	0.210 - 0.340
Atlanta	2.03	2	0.050 - 0.055	391 - 393	122 - 134	54 - 59	22 - 23	0.175 - 0.195
New Orleans	1.19	4	0.034 - 0.047	77 - 310	0 - 21	0 - 3	0 - 0	0.005 - 0.124
Memphis	0.91	2	0.044 - 0.051	294 - 535	23 - 41	6 - 11	1 - 2	0.145 - 0.150
Louisville	0.91	3	0.035 - 0.059	354 - 512	47 - 167	7 - 59	0 - 16	0.133 - 0.190
Nashville	0.85	3	0.029 - 0.044	79 - 343	0 - 60	0 - 5	0 - 0	0.095 - 0.136
Birmingham	0.85	3	0.049 - 0.058	281 - 420	29 - 57	10 - 24	0 - 6	0.136 - 0.171
Greensboro	0.83	2	0.050 - 0.052	297 - 299	10 - 19	0 - 2	0 - 1	0.113 - 0.143
Norfolk	0.81	2	0.052 - 0.063	445 - 564	74 - 97	10 - 16	0 - 3	0.135 - 0.154
Jacksonville	0.74	2	0.039 - 0.042	70 - 108	1 - 13	0 - 4	0 - 1	0.110 - 0.140
Orlando	0.70	2	0.036 - 0.037	61 - 94	0 - 7	0 - 3	0 - 0	0.095 - 0.130
Charlotte	0.64	3	0.052 - 0.058	316 - 536	26 - 133	2 - 20	0 - 4	0.131 - 0.155
Richmond	0.63	3	0.058 - 0.064	696 - 715	81 - 118	9 - 24	0 - 3	0.124 - 0.150
Greenville	0.57	1	0.052 -	425 -	25 -	0 -	0 -	0.117 -
Raleigh	0.53	2	0.053 - 0.054	407 - 576	37 - 55	4 - 8	0 - 2	0.130 - 0.143
Baton Rouge	0.49	4	0.031 - 0.043	54 - 226	8 - 38	2 - 5	0 - 1	0.130 - 0.169
Knoxville	0.48	2	0.032 - 0.045	58 - 376	1 - 19	0 - 0	0 - 0	0.102 - 0.113
Mobile	0.44	2	0.046 - 0.049	179 - 290	0 - 27	0 - 4	0 - 2	0.099 - 0.157
Kingsport	0.43	2	0.041 - 0.051	248 - 328	8 - 23	0 - 3	0 - 0	0.106 - 0.130
Charleston	0.43	2	0.038 - 0.045	64 - 112	2 - 3	0 - 0	0 - 0	0.112 - 0.118
Chattanooga	0.43	2	0.053 - 0.057	482 - 578	35 - 40	4 - 7	0 - 0	0.135 - 0.135
Columbia	0.41	2	0.052 - 0.052	231 - 405	13 - 16	0 - 3	0 - 0	0.119 - 0.126
Little Rock	0.39	2	0.044 - 0.049	261 - 393	2 - 12	0 - 0	0 - -	0.101 - 0.110
Shreveport	0.38	1	0.036 -	76 -	3 -	1 -	0 -	0.124 -
Beaumont	0.38	2	0.046 - 0.055	276 - 384	25 - 69	5 - 18	0 - 6	0.130 - 0.170
Newport News	0.36	1	0.052 -	463 -	56 -	9 -	0 -	0.136 -
Jackson	0.32	1	0.046 -	153 -	9 -	1 -	0 -	0.120 -
Lexington	0.32	3	0.041 - 0.051	260 - 523	7 - 58	0 - 6	0 - 0	0.111 - 0.137

Source: Pinkerton and Lefohn (1986).

of ozone from the western Gulf Coast area to the midwestern and the northeastern States has been observed (Wolff and Lioy 1980). Under similar meteorological conditions, transport of high concentrations of ozone from the New York, Philadelphia, and Washington, DC, corridor into Virginia, West Virginia, western Pennsylvania, and Ohio has been reported (Fankhauser 1976).

For all of the above reasons, transport of ozone and other pollutants from urban and industrialized areas to near-urban, rural, and even high-elevation forests is the most important factor determining ozone exposure of forests throughout eastern North America. The highest ozone concentrations occur in summer and fall when sunlight is most intense and stagnant meteorological conditions augment the conditions necessary for formation and accumulation of ozone.

e) Diurnal variability. The patterns of ozone occurrence in ambient air depend on daily and seasonal variations in the concentrations of precursor chemicals in the air, the ambient temperature, and the intensity of sunlight. In most urban areas in eastern North America, the typical diurnal pattern (observed at urban, near-urban, and rural monitoring sites) has a minimum ozone concentration near zero. This occurs around sunrise, then increases through the morning to an early afternoon peak concentration, and finally decreases toward minimal concentration again in the evening (fig. 18). Thus, high concentrations of ozone may occur for 6 to 10 hours per day.

An important variation on this basic diurnal theme occurs when a secondary peak appears in addition to the primary early afternoon peak. This secondary peak occurs only in some localities. It may occur any time from mid-afternoon to the middle of the night. It is attributed to ozone transported from an upwind area where high concentrations accumulated earlier in the day. Secondary peak concentrations may be higher than those in the primary peak. For example, at one rural site in Massachusetts, primary peak concentrations of approximately 110 ppb, 140 ppb, and 140 ppb occurred at noon, or from noon to 4 p.m., respectively, on 3 successive days. Secondary peaks at those same sites for the same 3 days were 150 ppb, 157 ppb, and 130 ppb; these occurred at 6:00 p.m., 8:00 p.m., and 8:00 p.m., respectively (fig. 19) (U.S. Environmental Protection Agency 1986).

High-elevation forests often experience no important diurnal variability. This is in distinct contrast to the pattern in low-altitude forests. One study at a rural site in New Jersey characterized ozone concentrations along an elevation gradient (see fig. 20) (Wolff and others 1987). Peak ozone concentrations were similar at each elevation, but the lower concentrations increased with elevation. Ozone concentrations at this site varied during the day by as much as 100 ppb.

f) Daily to weekly variability. During episodes of high ozone concentration, diurnal patterns may vary from day to day with peaks occurring at any time during the day or night. At many monitoring sites throughout eastern North America,

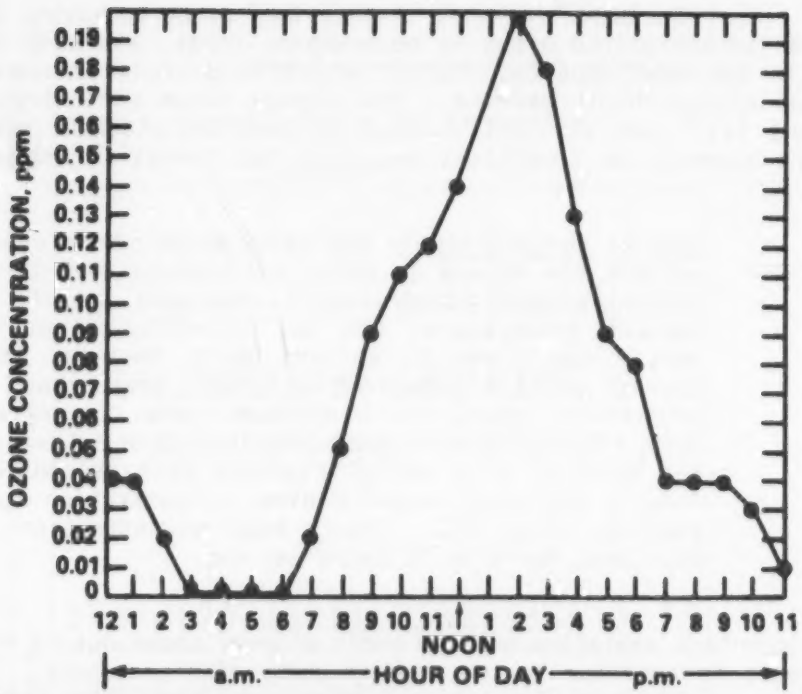


Figure 18. Diurnal pattern of 1-hour ozone concentrations on July 13, 1979, Philadelphia, PA.

Source: SAROAD (1985).

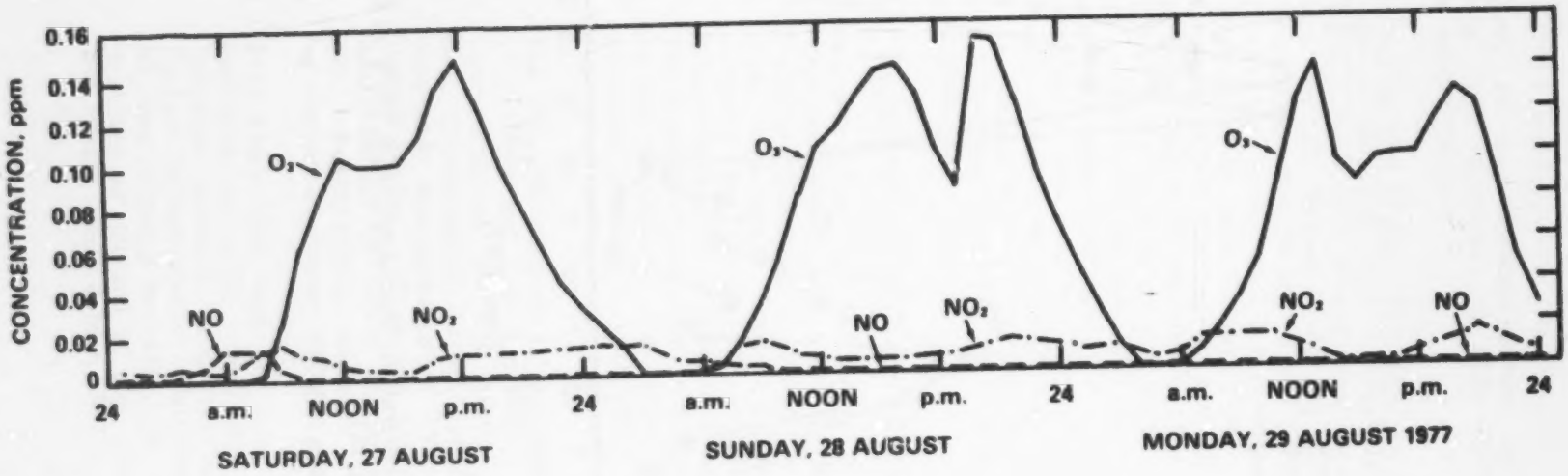


Figure 19. Three-day sequence of hourly ozone concentrations at Montague, MA, SURE station showing locally generated midday peaks and transported late peaks.

Source: U.S. Environmental Protection Agency (1986).

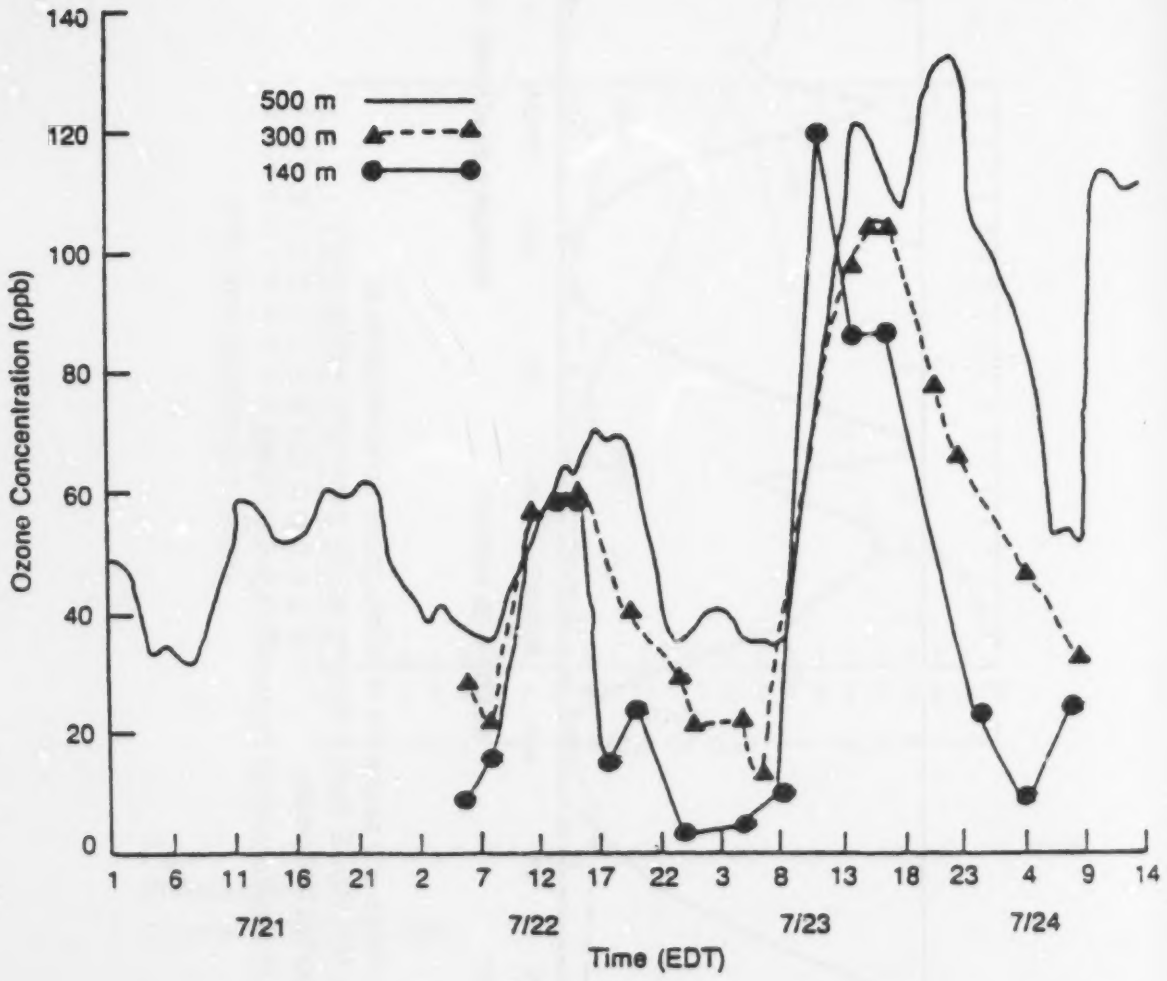


Figure 20. Ozone concentrations from July 21–24, 1975, at three altitudes at High Point, NJ.

Source: Wolff and others (1987).

episodes of high concentrations of ozone occur more frequently than episodes of high sulfur dioxide or nitrogen oxide concentrations.

4) Co-occurring and sequential exposures to sulfur dioxide, nitrogen oxides, and ozone: Possible co-occurrences of SO_2, NOx, and O_3 have been investigated at 30 monitoring sites in the Eastern United States. During 1978-81, co-occurrences of concentrations of > 50 ppb of all three pollutants were observed very rarely, i.e., < 1.5 percent of the monitoring hours analyzed (Lefohn and Tingey 1984). This was also true for co-occurrences of NOx and O_3 and NOx and SO_2. But SO_2 and O_3 co-occurred about 30 percent of the time. Sulfur dioxide and O_3 co-occurred because O_3 was very often present at concentrations greater than 50 ppb, and SO_2 concentrations greater than 50 ppb occurred periodically (Lefohn and Tingey 1984). Scavenging of NOx during the formation of ozone explains why NOx and ozone rarely occur together--NOx concentrations are usually lowest during the day when ozone concentrations are usually highest (see fig. 19) (U.S. Environmental Protection Agency 1986).

5) PAN, PPN, hydrogen peroxide, and nitric acid:

a) Concentrations of PAN, PPN, hydrogen peroxide, ammonia, nitrogen oxides, and other oxidant gases usually are lower than concentrations of ozone and sulfur dioxide but in some areas may equal them. Concentrations of these other oxidants including PAN and PPN are usually below the detection limits of most available monitoring instruments. The reason for considering these pollutants in this chapter is the possibility that they co-occur with other airborne pollutant chemicals with which they may cause additive or synergistic effects on vegetation.

b) PAN and PPN concentrations typically occur at about 4 to 12 percent and 1 to 2 percent, respectively, of the ozone concentrations at any given location (Altshuller 1986; U.S. Environmental Protection Agency 1986). There are no available measurements of PAN or PPN for the Southeastern United States. If we use the above percentages to convert measurements of ozone concentration to estimate PAN and PPN concentrations however, PAN and PPN in combination may occur at 7-hour average concentrations of 8 to 21 ppb.

c) Only since 1984 have gas-phase concentrations of H_2O_2 been measured reliably. Available data suggest that maximum hydrogen peroxide concentrations occur in summer in the Eastern United States. Measurements at Whiteface Mountain, NY, and Whitetop Mountain, VA, indicate 1-hour peak concentrations of 60 and 80 ppb, respectively (fig. 21) (Heikes and others 1987; Mohnen 1987).

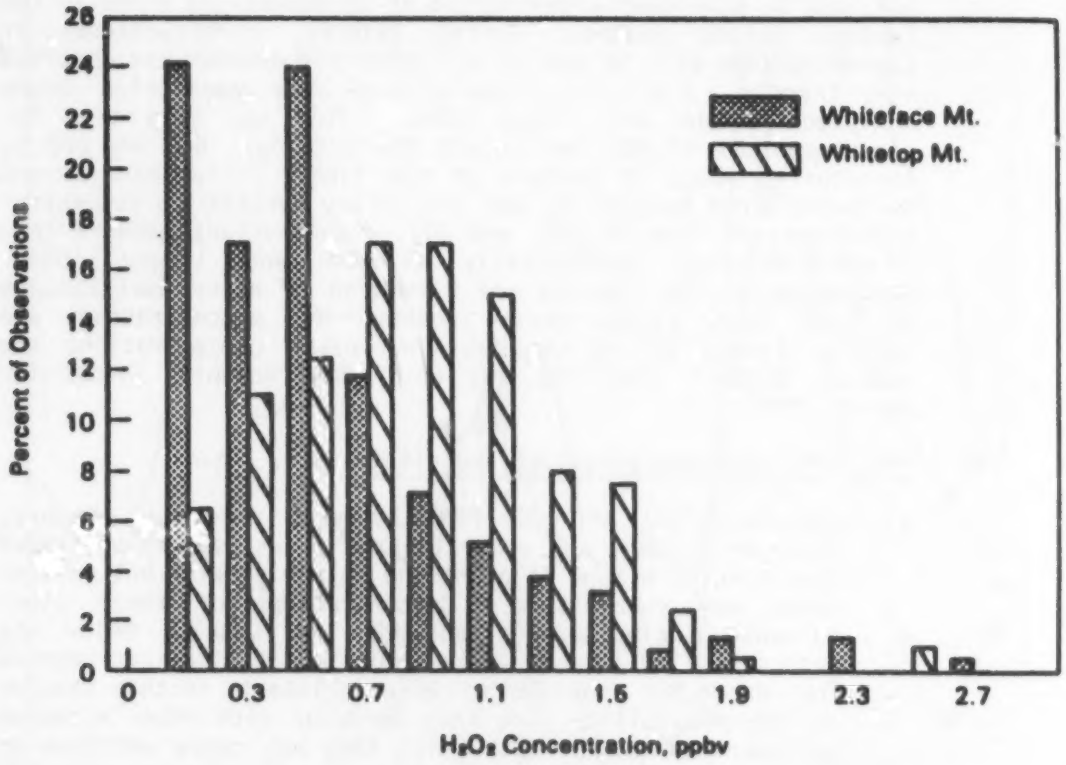


Figure 21. Distribution of hourly, summertime concentrations at two high elevation sites.

Source: Heikes and others (1987).

d) Only very sparse data for ammonia and nitric acid exposure are available. Measurements of ammonia and nitric acid at three southeastern sites averaged approximately 1 to 3 ppb and <1 ppb, respectively; in many other studies, concentrations were usually below the detection limits for the instruments used (Cadle and others 1982; Kelly and others 1984). Possible co-occurrence of these compounds with other airborne pollutant chemicals cannot be described from these few measurements.

6) Acid deposition: The acidity of precipitation and cloud water (hydrogen ion concentration, usually expressed as pH) is determined by the balance among all the airborne cations and anions dissolved in these components of wet deposition. Similarly, the acidifying effects on forest ecosystems of all the airborne gases, aerosols, and dissolved and suspended substances in precipitation and cloud water are determined by the balance among the airborne cations and anions transferred from the atmosphere, and the exchange and transformation processes that take place with the above- and below-ground parts of the plants and soils of the forest. Thus, the principal acidity determining ions include K^+, Na^+, NH_4^+, Ca^{++}, Mg^{++}, Cl^-, NO_3^-, $SO_4^=$, $PO_4^{\equiv}$. Among these, ammonium and hydrogen are the most important cations, while sulfate, nitrate, and occasionally chloride are the most important anions. Ammonium ion has a distinctive role since it is neutralizing in its effects in precipitation and cloud water but is acidifying in its effects when taken up by plants, animals, and microorganisms in forest ecosystems.

a) Total loading. The total loading of sulfate, nitrate, ammonium, and hydrogen ions into forest ecosystems is determined by the wet and dry deposition of five types of substances, the first four of which undergo conversion and dissolution processes as soon as they contact the moist surfaces of plants, animals, microorganisms, soils, rocks, and water films, droplets, and pools:

- Gaseous sulfur dioxide is further oxidized to sulfate ions in solution;
- Gaseous nitrogen oxides (NO_2 and NO), PAN, PPN, and nitric acid vapor are all further oxidized to nitrate ions in solution;
- Gaseous ammonia is first dissolved in water and then dissociated into ammonium ions in solution;
- Airborne aqueous or dry aerosols (which consist mainly of ammonium nitrate, ammonium sulfate, and ammonium acid sulfate) dissolve and then dissociate into a mixture of sulfate, nitrate, ammonium, and hydrogen ions in solution; and all of the above are mixed with
- cloud water and precipitation that already contain a mixture of sulfate, nitrate, ammonium, and hydrogen ions in solution.

If the gases and aerosols listed above are deposited on truly moisture-free surfaces, it will still be only a short time (usually only a few hours or days) before these surfaces will be wetted by dew, by precipitation, or by fog or cloud water. Thus, the end-products of all these dissolution, dissociation, and mixing reactions will be incorporated into the soil solution or into the moisture that bathes the above-ground portions of the plants, animals, microorganisms, soil particles, and rocks of forest ecosystems. The sulfate, nitrate, ammonium, hydrogen, and other cations and anions in these solutions will then be free to diffuse:

- through stomata, cuticle, cell walls, and cell membranes on their way into the cytoplasm of leaves of trees and other forest plants, or

- into the soil pores, soil colloids, cell walls, cell membranes, and on into the cytoplasm of the fine feeder roots or mycorrhizae of forest vegetation.

b) Regional variability in wet deposition of sulfate, nitrate, ammonium, and hydrogen ions. Since 1980, direct measurements of the concentrations and deposition of the major cations and anions in precipitation are made weekly at wet-deposition monitoring sites in various parts of North America (NADP 1985). At present there are about 200 such sites in the United States and about 60 in Canada. Thus, reliable maps are now available describing the geographical, seasonal, and multi-year variability in the concentration and deposition of sulfate, nitrate, ammonium, hydrogen, and other ions in precipitation (see figs. 22 through 25). Year-to-year variability in these maps for both concentration and deposition is fairly small; thus, the maps shown in these figures for 1985 are representative of the concentration and deposition of these ions in precipitation in various parts of eastern North America in the years between 1980 and 1986.

These maps show the following regional trends in concentration and deposition in precipitation across eastern North America:

- Sulfate concentration in precipitation varied from a maximum of about 3 milligrams per liter in portions of Ohio, Pennsylvania, and Ontario and decreased progressively to less than 1 mg/L in portions of Quebec and Ontario to the north and North Dakota and Texas to the west, and to between 1 and 1.5 milligrams per liter across the Southeastern States (see fig. 22a).

- Sulfate deposition in precipitation varied from a maximum of about 35 kilograms per hectare in northern Ohio, and decreased progressively to less than 10 kilograms per hectare in portions of Quebec and Ontario to the north and the States between North Dakota and Texas to the west, and to between 10 and 20 kilograms per hectare across the southern States (see fig. 22b).

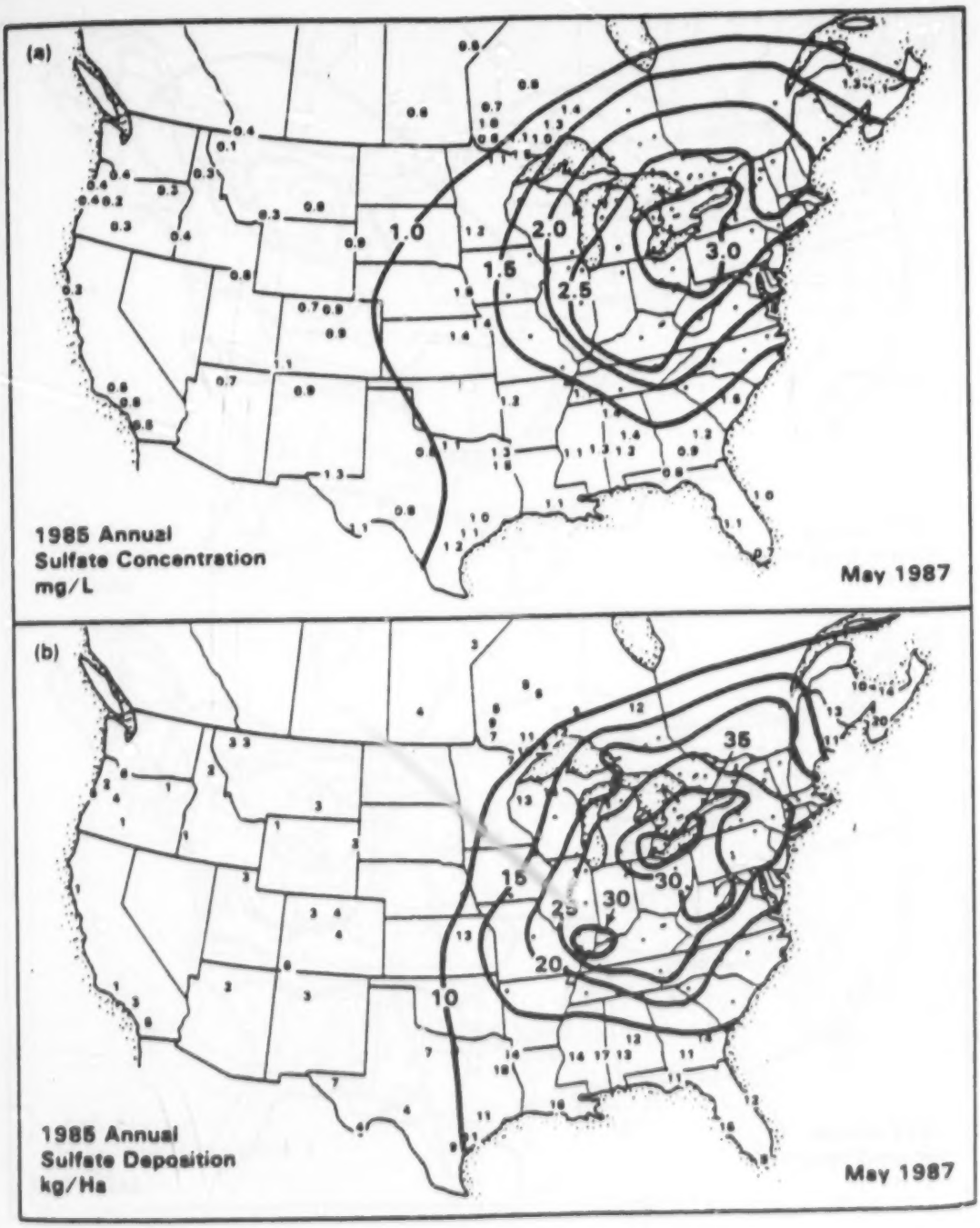


Figure 22. The 1985 annual distribution of SO_4^{2-}: (a) concentration and (b) deposition. Contour intervals are (a) 0.5 mg/L and (b) 0.5 g/m^2.

Source: Barchet (1987).

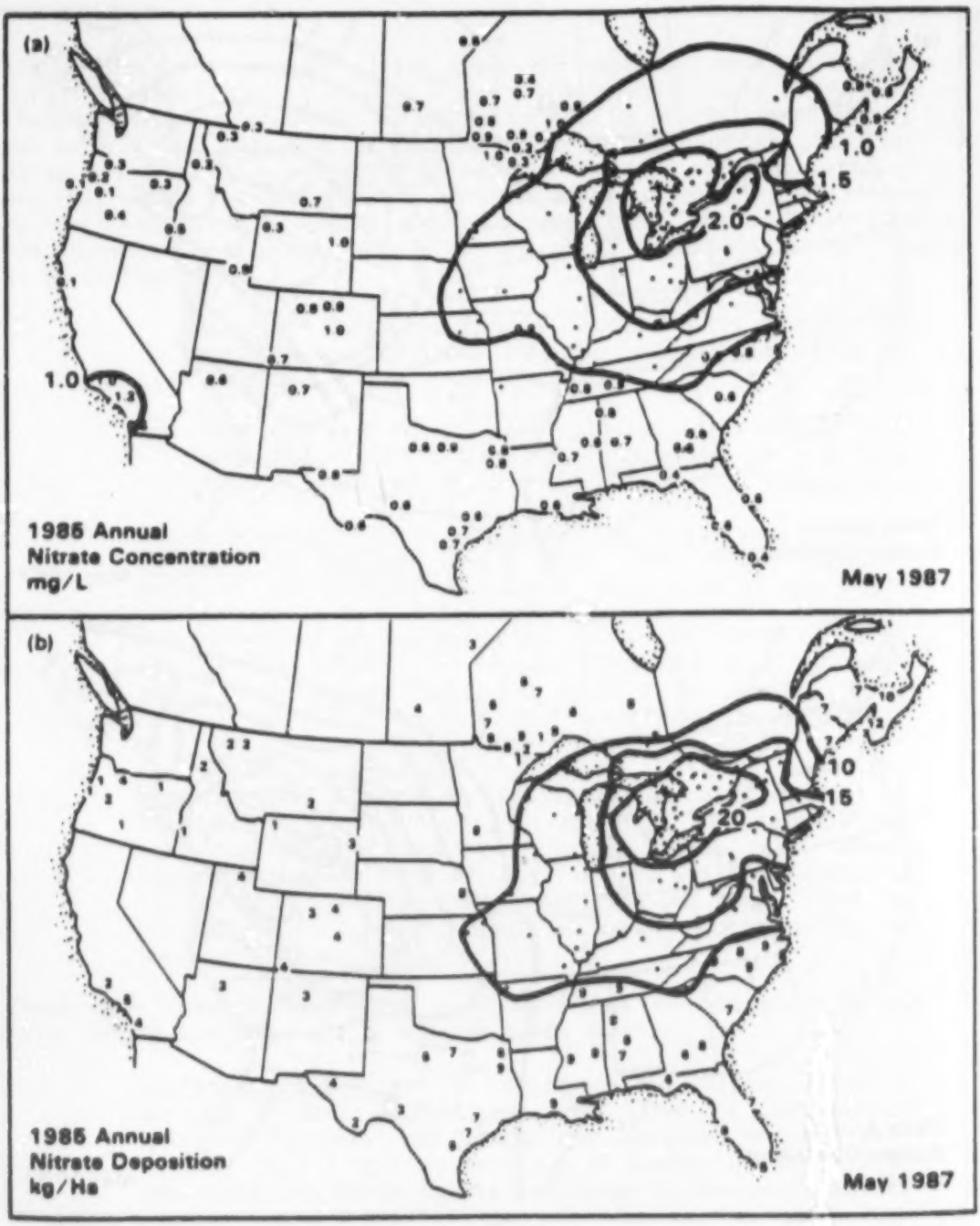


Figure 23. The 1985 annual distribution of NO_3^-: (a) concentration and (b) deposition contour intervals are (a) 0.5 mg/L and (b) 0.5 g/m^2.

Source: Barchet (1987).

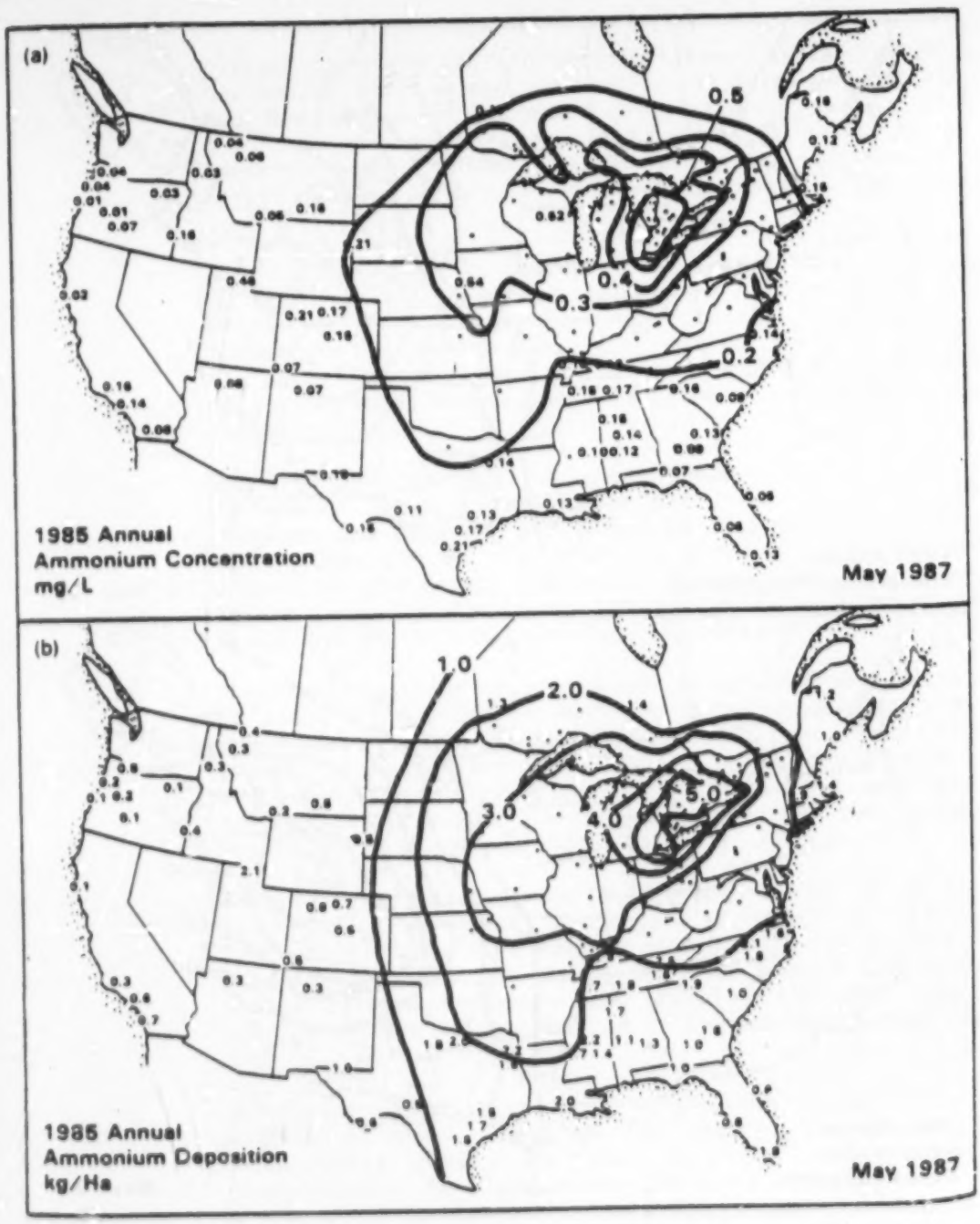


Figure 24. The 1985 annual distribution of NH_4^+: (a) concentration and (b) deposition. Contour intervals are (a) (0.1 mg/L and (b) 0.1 g/m^2.

Source: Barchet (1987).

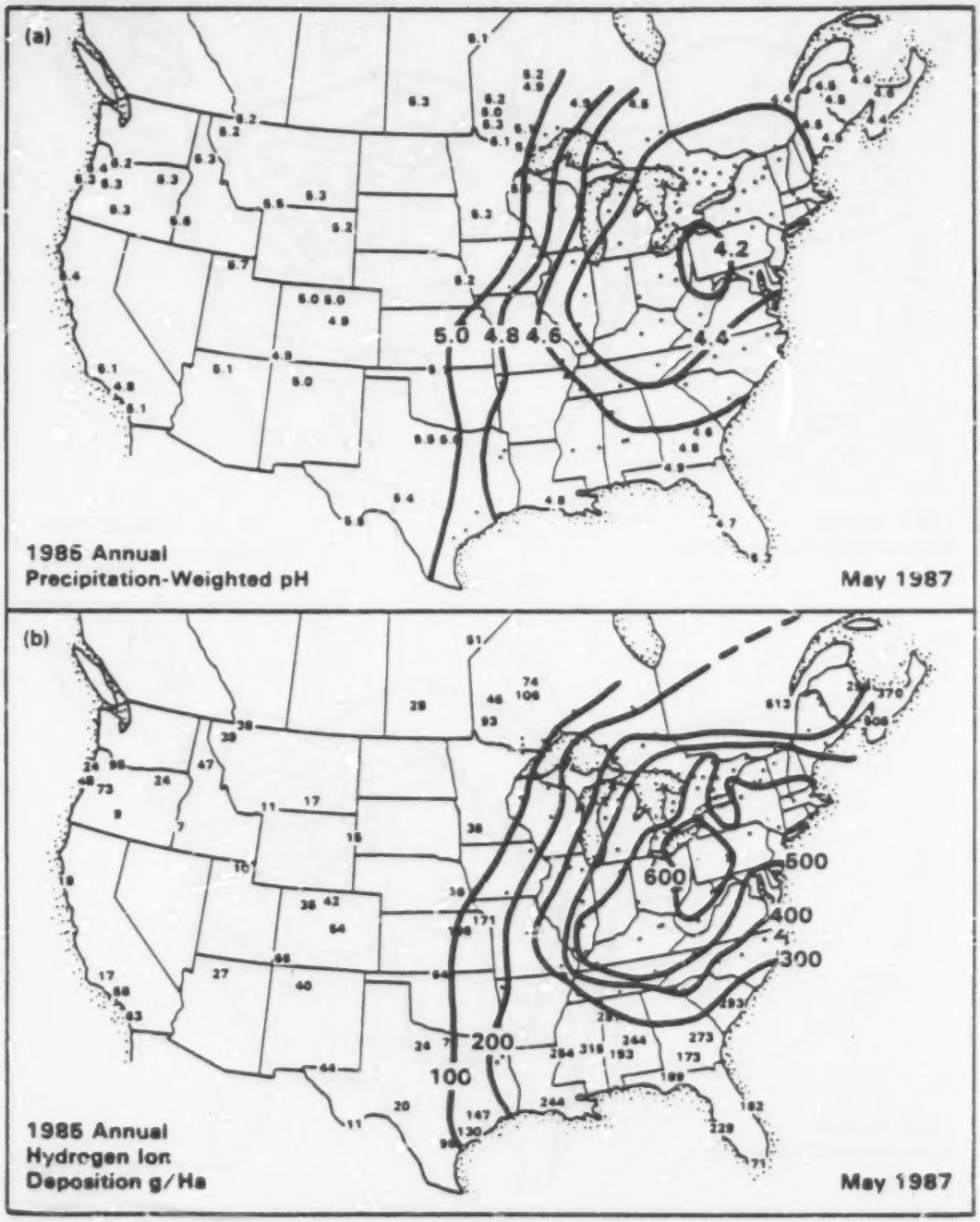


Figure 25. The 1985 annual distribution of (a) precipitation – weighted pH and (b) H^+ deposition. Contour intervals are (a) 0.2 pH units and (b) 20 mg/m^2.

Source: Barchet (1987).

- Nitrate concentration in precipitation varied from a maximum of about 2 mg/L in portions of Michigan, Ontario, and New York, and decreased progressively to less than 1 mg/L in portions of Quebec and Ontario to the north, the States between Minnesota and Kansas to the west, and the States between Arkansas and North Carolina to the south (see fig. 23a).

- Nitrate deposition in precipitation varied from a maximum of about 20 kilograms per hectare in northern Ohio, and decreased progressively to less than 10 kilograms per hectare in portions of Quebec and Ontario to the north, the States between Minnesota and Oklahoma to the west, and the States between Arkansas and North Carolina to the south (see fig. 23b).

- Ammonium concentration in precipitation varied from a maximum of about 0.5 milligrams per liter in portions of Michigan and Ontario, and decreased progressively to less than 0.2 milligrams per liter in portions of Quebec and Ontario to the north, the States between North Dakota and Colorado to the west, and the States between Texas and North Carolina to the south (see fig. 24a).

- Ammonium deposition in precipitation varied from a maximum of about 5 kilograms per hectare in portions of Ohio and Ontario, and decreased progressively to less than 1 kilogram per hectare in portions of Quebec and Ontario to the north, the States between North Dakota and west Texas to the west, and to less than 2 kilograms per hectare across the Southeastern States (see fig. 24b).

- The volume-weighted average pH of precipitation varied from a minimum of about 4.2 in portions of Pennsylvania, Ohio, and West Virginia to a maximum of above 4.5 in portions of Ontario and Quebec to the north, a maximum of 5.0 in the States between Minnesota and Texas to the west, and a maximum between 4.4 and 4.8 across the southern States (see fig. 25a).

- Hydrogen ion deposition in precipitation varied from a maximum of about 600 grams per hectare in portions of Pennsylvania, Ohio, and West Virginia, and decreased progressively to less than 200 grams per hectare in portions of Quebec and Ontario to the north, less than 100 grams per hectare in the States between Minnesota and Texas to the west, and to between 200 and 400 grams per hectare across the southern States (see fig. 25b).

In general, sulfate concentrations are greater in summer than in winter. In a broad band extending from New England southwestward along both the eastern and western sides of the Appalachian Mountains as far as western North

Carolina, the ratio of summertime to wintertime sulfate concentration is greater than two to one.

Wet deposition of sulfate in precipitation in warm periods is highly episodic -- 50 to 70 percent of the total annual deposition of sulfate occurs in about 20 percent of the total days (Summers and others 1986).

Wet deposition of nitrate ions in precipitation is also highly episodic -- 50 to 70 percent of the total annual deposition of nitrate occurs on about 20 percent of the total days per year. High nitrate deposition occurs more often in warm than in cool periods, but does not show as strong seasonal dependence as sulfate deposition (Summers and others 1986).

Sulfate and nitrate concentrations in cloudwater have been measured at a few high-elevation sites in the northeastern States and in Virginia. In 1984, sulfate concentrations ranged from about 930 to 1850 microequivalents per liter, while nitrate concentrations in 1984 ranged from about 190 to 900 microequivalents per liter (Weathers and others 1986). High elevation forests in the northern and southern Appalachian Mountains are immersed in clouds on more than half the total days per year (Mohnen 1987).

The pH of cloudwater in both the northeastern and southeastern States typically averaged 3.5, with highly acidic events of around pH 2.6 (Mohnen 1987; Weathers and others 1986).

c) Regional variability in dry deposition of sulfate, nitrate, ammonium, and hydrogen ions. In contrast to the situation for wet deposition, no reliable measurements of regional variability in dry deposition of gases and aerosols leading to the accumulation of sulfate, nitrate, ammonium, and hydrogen ions in forest ecosystems have been made in North America. The fragmentary information that is available is based mainly on short-term studies in a few specific locations and on ambient air quality monitors established mainly in urban areas. The amounts of gases and aerosols present in the atmosphere of most rural, high-elevation remote, and even many near-urban locations are so low that they are at or below the detection limits for the monitors that are used. These instruments were designed and installed mainly for the purpose of insuring compliance with human-health-related air quality monitoring in cities. Thus, these data cannot be used to give reliable estimates of air concentrations or dry deposition of sulfate, nitrate, ammonium, and hydrogen ions.

The only direct measurements of the regional distribution of gaseous sulfur dioxide and sulfate aerosol were made in 1977-78 during the Sulfate Regional Experiment conducted by the Electric Power Research Institute. As shown in figure 15:

- Gaseous sulfur dioxide concentrations varied from an arithmetic 1-hour average of about 20 ppb in portions of West Virginia and Pennsylvania, to less than 10 ppb in Ontario and New England to the north, portions of Michigan and Illinois to the west, and almost all of the southern States to the south (see fig. 15a);

- Sulfate aerosol concentrations decreased from a maximum geometric 24-hour average of about 8 micrograms per cubic meter in portions of Alabama, Kentucky, and Pennsylvania, to a minimum of less than 4 micrograms per cubic meter in New England and Ontario to the north, the States between Wisconsin and Missouri to the west, and to less than 6 micrograms per cubic meter in the Gulf and Atlantic Coast States (see figure 15b) (Mueller and Hidy 1983).

Thus, sulfate aerosol is more uniformly distributed, on the average, than sulfur dioxide, its gaseous precursor. The more uniform distribution results from the dispersion, mixing, and long-distance transport of sulfur dioxide during its conversion to sulfate (Mueller and Hidy 1983). High concentrations of sulfate aerosol frequently accumulate in stagnant air masses as sulfur dioxide is converted to sulfate aerosol. These high sulfate episodes sometimes persist for several days and extend over large areas of eastern North America (Mueller and Hidy 1983).

Unfortunately, similarly reliable data for gaseous nitrogen oxides and nitrate aerosol are not available.

In theory, it should be possible to calculate the amounts of sulfate, nitrate, ammonium, and hydrogen ions transferred from the atmosphere into forest ecosystems in various regions of North America by multiplying the values for regional measurements of the air concentration of gases and aerosols by the highest and lowest reasonable estimates of the deposition velocity for these substances. Such broad regional estimates of dry deposition of gases and aerosols have not found wide acceptance, however (Hicks 1984).

4. EFFECTS OF AIRBORNE SULFUR- AND NITROGREN-DERIVED POLLUTANT CHEMICALS ON FOREST ECOSYSTEMS OF EASTERN NORTH AMERICA

4.1 How do Forest Ecosystems Respond to Changes in the Physical and Chemical Climate?

Mature forest ecosystems are seldom stable, therefore, assessing ecosystem responses to stresses is extremely difficult. They maintain themselves in an oscillating steady state characterized by the continuous elimination of suppressed and old trees and the addition of new ones. Intense competition among trees for light, water, nutrients, and space, as well as recurrent natural climatic and biological stresses (such as those cited below), alter the species composition of mature forests by eliminating those individuals sensitive to specific stresses. Competition among trees of the same species does not affect the composition of stands and does not influence species succession. Competition among different species, however, results in succession and ultimately produces a mature forest with forest trees and other plant species having a capacity to tolerate the competitional stresses (Kozlowski 1980; Kozlowski and Constantinidou 1986a). Succession, unless there is a catastrophic disturbance, can take 100 to 1000 years.

In a mature forest, a mild disturbance has little effect on its oscillating steady state. However, if injury or disturbance, whether from air pollution or other stresses, leads to changes in the species composition of the forest severe enough to disrupt food chains and modify the rates of energy flow and nutrient cycling, succession may be returned to an earlier, less complex, successional stage (Bormann 1985; Woodwell 1970).

Ecosystems respond to changes in their environment through the organisms that comprise them. Forest ecosystem responses to stress begin with the interaction of the individual organism with its environment. Three levels of interaction are involved: (1) between the individual tree and its environment; (2) between the population and its environment; and (3) between the forest (the biological community composed of many species) and its environment (Billings 1978).

In most ecosystems, the principal interaction among species is competition for resources. In their struggle for existence, forest trees are continually undergoing one or more of the following natural environmental stresses (Woodman and Cowling 1987):

1. Natural competitional stresses. These occur whenever trees compete with each other or with other plants for limited supplies of growing space, solar radiation, water and/or essential nutrients from the same soil or microclimate. Competitional stresses are the dominant cause of low vigor, poor growth, and death of trees in most forests. More trees die of competitional stresses than from all other stresses combined (Woodman and Cowling 1987);

2. Natural climate stresses. These include extremes of high and low temperature, drought, flooding, low humidity, and high winds. Water stress is the most common cause of decreased growth, low vigor, premature loss of foliage, and mortality (Kramer 1983). Temperature and the availability of water have the greatest impact on growth in the northern hemisphere (Osmond and others 1987);

3. Natural biological stresses. These include parasitic and pathogenic fungi, insects, nematodes, bacteria, viruses, viroids, plasmids, mycoplasmas, parasitic seed plants, and herbivorous animals such as porcupines. In forests, fungi and insects are the most important biotic pathogens. Any of these biotic stresses can cause impairment of normal physiological processes. The effects of these biotic agents vary over time because their activities depend upon the environmental requirements of the predators and pathogens (Osmond and others 1987);

4. Natural chemical stresses. These are induced by deficiencies (and occasionally excesses) of essential nutrients or toxic soil chemicals such as aluminum. Nitrogen is the nutrient most commonly deficient in forests. Virtually all of the nitrogen and sulfur entering a forest ecosystem comes from the atmosphere (Waring and Schlesinger 1985).

Changes that occur in the physical or chemical climate of plant populations apply new selection pressures (Treshow 1980a; Woodwell 1970). The subtle and indirect effects of pollutant dosages can set the stage for changes in community structure that may possibly have irreversible consequences (Guderian and Kueppers 1980). Increasing pollutant stress provides a selective force that favors some genotypes, suppresses others, and eliminates those species that lack sufficient genetic diversity to survive. Thus, the occurrence and distribution of plants is influenced, and community composition and species interactions are changed such that the basic structure of the ecosystem is ultimately changed (Treshow 1980a). This succession may take years, decades, or longer, depending on the pollutant concentration, period of exposure, and plant species involved.

Man-made stresses that change the chemical and physical climates of plants are of two main types:

1. Air pollution stresses. These are chemical stresses that occur whenever forest trees are exposed to injurious concentrations of toxic gases such as sulfur or nitrogen oxides, ozone, or fluoride; toxic aerosol particles or coarse particulate matter; or dissolved or suspended chemicals in air, cloud water, or precipitation. Pollutant stresses can also occur as a result of accumulation or mobilization of toxic substances in soils often after wet or dry deposition from the atmosphere (Woodman and Cowling 1987).

2. Mechanical stresses. These result from various human activities including logging, controlled burning, draining, flooding, and physical disturbance of soils leading to compaction, erosion, leaching of nutrients, or accumulation of toxic substances (Woodman and Cowling 1987).

The above stresses, both natural and anthropogenic, can occur at the same time or in any order, one after another. They can act independently, additively, synergistically, or antagonistically; they can be periodic, either short- or long-term, and can increase or decrease with the age of the forest stand (Cowling 1985).

Air pollution stresses generally result not because gases and chemicals are unique substances never before encountered by plants, but rather because the amounts of these substances are beyond the limits of tolerance or avoidance of the organism. Acute air pollution stress tends to be episodic. Trees experience high pollutant exposures for minutes, hours, or for a few days to a week at the most. These acute exposures may occur several times during a year. In chronic air pollution stress, however, low concentrations are experienced continuously for a considerable portion of the life of the plant.

In theory, airborne chemicals in the form of gases or aerosols, when suspended in air or dissolved in cloud water or precipitation, could cause, either alone or with other stress factors, many different types of effects in forests. These include the following:

Effects on Individual Trees:

1) Visible symptoms of injury to individual trees-

- Change in shape and size of leaves,
- Change in normal patterns of growth and development (shape and timing of development of leaves, flowers, buds, branches, roots, etc.),
- Change in normal senescence patterns of leaves, self-pruning of branches, or turn-over of feeder roots,
- Change in life expectancy of individual trees;

2) Decreased annual height, growth, or radial increments (with or without visible symptoms of injury to individual trees);

3) Alteration of physiological processes (with or without visible symptoms)-

- Photosynthesis,
- Respiration,
- Transpiration,
- Mineral nutrition,
- Transport and allocation of photosynthate,
- Reproduction,
- Hormonal control of growth,
- Symbiotic relationships with other organisms;

4) Changes in susceptibility to other stress factors;

5) Changes in reproductive behavior of individual trees;

Effects on Forest Stands:

6) Decreased productivity of whole forest stands;

7) Change in stand stocking, such as age-class distribution;

8) Changes in normal patterns of competition and mortality within forest stands (both among individual trees and between trees and other vegetation in forest stands);

9) Changes in normal patterns of succession within forest stands;

10) Changes in species composition of forest;

11) Changes in nutrient cycling in forest stands (such as excessive leaching of nitrate or other mobile anions or cations from soils);

12) Changes in hydrologic behavior and watershed functions of forest stands;

Effects on Tree Species:

13) Changes in the genetic diversity within tree species (alteration of gene pools within tree species);

14) Changes in fitness within and between tree species;

Effects on Regional and Social Values of Forests:

15) Changes in the regional productivity, distribution, or economic value of forests; and

16) Changes in aesthetic quality or other ecosystem values for humans.

Fifteen of these 16 theoretical effects have been confirmed by observation and experiments in forests of North America. In both North America and Europe, studies of the effects of air pollutants on forests and agricultural systems have been made in the vicinity of strong point sources of sulfur dioxide (Legge and others 1981; Linzon 1978; Miller and McBride 1975; Scheffer and Hedgcock 1955; Winner and Bewley 1978a,b; Winner and others 1978); fluoride, (Miller and McBride 1975; Treshow and others 1967); and chronic radiation (Woodwell 1970). Much less is known about the effects of regionally dispersed airborne chemicals. Ozone has been the most studied regional pollutant (Miller 1973; Miller and others 1982). Only number 15 listed above has not been associated with regional ozone exposure in the San Bernardino Forest of California.

Air pollutants may affect trees directly through the foliage or indirectly via the soil. Regardless of whether the air pollution stress is acute or episodic, forest trees or perennial shrubs must cope with the cumulative effects of both short- and long-term stresses. Tree response may appear rapidly as when, for example, sensitive white pine needles show visible injury from exposure to high concentrations of ozone. In other instances, however, responses are often subtle and may not be observable for many years because adaptation and response to stress is expressed by the differential growth that results from changes in carbon allocation (Waring and Schlesinger 1985).

Tree populations play a critical role in mature forest ecosystems. They represent the later stages of succession and are adapted for high competitive ability (Brown 1984). As the dominant producers, trees influence the structure (species composition and trophic relationships), energy flow, and nutrient relationships (Ehrlich and Mooney 1983). The ecosystem processes of energy flow and nutrient cycling are directly related to the plant physiological processes of photosynthesis, nutrient uptake, biosynthesis, carbon allocation, respiration, and translocation. The alteration of these physiological processes is the fundamental cause of all other ecosystem effects (U.S. Environmental Protection Agency 1986).

Changes in ecosystems begin with the death of individual plants. Temporal dynamics at the level of individual organisms can, therefore, upset the equilibrium and be disorganizing in ecosystems such as forests that are dominated by nonmobile organisms (Shugart 1987). In forests with large canopy trees, a canopy tree dominates the space where it is growing, reduces the amount of light reaching the forest floor and alters the microclimate, conditions that help to determine the plant species that can survive beneath the canopy. Death of a tree opens the canopy and changes the microclimate and substantially increases the resources of light, nutrients, water and energy available to other organisms. This change initiates a struggle for dominance among the understory trees and seedlings. In time a new canopy becomes established (Shugart 1987).

The mode of death of a tree is also ecologically important as it determines the regeneration success of trees that form the next forest generation. Trees may die catastrophically as when high winds or ice storms break off the crown or branches or when they are blown over, exposing the roots; or they may die slowly and tend to waste away as in the case of those injured by pathogens or insects (Shugart 1987). Tree death, as influenced by gaseous air pollutants such as ozone, is usually gradual rather than catastrophic unless the tree is extremely sensitive or the pollutant concentration is extremely high. Growth responses require time. Therefore, because growth responses are the result of cumulative stresses and trees are continually being subjected to many other stresses, the cause of death is difficult to determine.

Pollutant stresses, as well as natural and biological stress as indicated above, have important implications for forest succession because they provide forces that favor some genotypes, affect others adversely, and eliminate species that lack genetic diversity (fig. 26). A population tolerant to the dominant stresses becomes established. However, studies using heavy metals and herbicides indicate that once the dominant stress is removed, the pollutant-tolerant plants tend to decline in number. Competition increases selection for

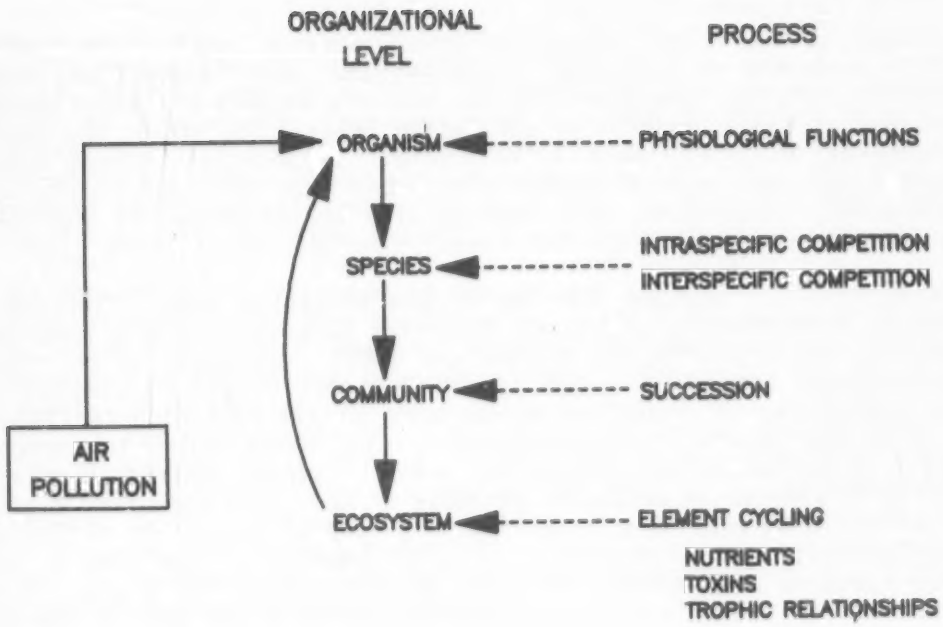


Figure 26. Pathways of air pollutant impact in forest ecosystems. Source: McClenahen (1984).

tolerance under polluted conditions and against tolerance under less polluted conditions (Roose and others 1982). This evidence is corroborated by observations of ecosystems functioning under natural conditions. Certain terrestrial ecosystems require major disturbances (e.g., fire, drought, windstorms) to retain their characteristics (Smith 1980; Vogl 1980). The development of tolerance depends upon the genotype being present in the plants when they are growing in environments free from air pollution or other selective stresses (Roose and others 1982).

Responses at the forest ecosystems level, because they are the summation of multiple responses of individual organisms and their interaction, usually take longer to develop, are more diffuse, and are usually of longer duration than responses at the individual or population level. Therefore, the cause and the response are more difficult to determine. In addition, forest stands differ greatly in age, species composition, stability, and capacity to recover from disturbance. Therefore, data dealing with the responses of one forest type may not be applicable to another forest type (Kozlowski 1980).

Explanations for changes observed at the ecosystem level often can be determined by understanding processes occurring at physiological, population, and community levels (Waring and Schlesinger 1985). Responses at the individual tree level are determined by the extent that physiological processes within the tree are altered or inhibited by exposure to air pollution (McClenahen 1984). It is now generally accepted that air pollution injury to individual organisms is initiated at the biochemical level. Changes induced in cells lead sequentially to changes in metabolic pathways of the whole organism, a disorganization of cellular membranes resulting in a loss of control of diffusion of water and solutes, and a breakdown of cellular components that finally results in visible symptoms such as chlorosis and necrosis of foliar tissue. Growth reduction, loss in yield (i.e., reproduction) and in plant quality can occur without visible injury to the plant (U.S. Environmental Protection Agency 1986; Kozlowski 1980).

Studies of the responses at the cellular level of trees most sensitive to specific stresses, both natural and anthropogenic, could lead to the identification of those physiological processes most likely to be affected by changes in the physical and chemical climate of forest ecosystems. Concentrations of the chemical compounds, the length of exposure, and the amount of the compound taken up are critical aspects of each individual plant response. Responses at the cellular level are determined in part by these factors. In the next section, the concepts of exposure, concentration, and uptake are discussed.

4.2 What is the Likelihood That Acid Deposition and Other Airborne Sulfur- and Nitrogen-Derived Chemicals Affect the Forests of Eastern North America Through Foliage-Mediated Response Mechanisms?

Plant response to air pollution stress involves pollutant concentration, length of exposure, deposition, uptake, and alteration of biochemical processes. Gaseous airborne chemicals (SO_2, NO_x, O_3, and mixtures of them) cause phytotoxicity of trees and other plants by entering the leaves and disrupting the biochemical processes that occur within the cells. The severity of injury usually is determined by the extent physiological processes are altered by the amount of pollutant entering the leaves. Plant response cannot be related solely to the concentration of the pollutant gas in the ambient air

(Unsworth and others 1976). The factors influencing chemical deposition, concentration, uptake, and the manifestation of injury are discussed in the following section. Plant response may be characterized by a measurable change in any plant function such as respiration, photosynthesis, rate of gas exchange, biochemical pathways, physiological reactions, degree of visible leaf injury, or decrease in growth or reproduction (yield) (U.S. Environmental Protection Agency 1986).

Visible injury to foliage does not necessarily mean that decreases in growth have occurred or are occurring. It is, however, an indication that phytotoxic concentrations are present and that studies should be conducted to assess the risk to vegetation from the concentrations present. Many studies, primarily of crop plants, have been made to assess the effect of a particular concentration and period of exposure. Table 9 presents the time period and concentration of three gaseous airborne chemicals (SO_2, NO_2, O_3) that have been reported to induce visible injury to vegetation in North America. Most of the data have been obtained from chamber, greenhouse, or field experiments, not from ambient exposures. The table may be used as a guideline to determine whether concentrations reported in the ambient air are likely to cause visible injury to vegetation or, more specifically, trees. Few exposure statistics are available for nitric acid, nitrate, ammonia, the ammonium ion, sulfate, hydrogen peroxide, or PPN (peroxy propionylnitrate) since these substances have not been widely identified as causing visible foliar injury. PAN (peroxyacetylnitrate), a regional pollutant in southern California, has not been widely reported in eastern North America. Though PAN is more toxic to crops than ozone, studies indicate trees are not as sensitive (U.S. Environmental Protection Agency 1986). Ambient air concentrations of nitrogen oxides (NO and NO_2) high enough to cause vegetational injury in the field have not been reported in eastern North America (National Research Council 1977b; U.S. Environmental Protection Agency 1982b).

Research has not yet clearly defined which components of pollutant exposure (pollutant concentration, time of exposure, time since previous exposure, etc.) are most important in causing plant responses. The characterization and representation of exposures to air pollutants has been and continues to be a major problem. An appropriate summary statistic for one exposure duration usually cannot be easily transformed to describe a different duration of exposure without access to the original aerometric data. In addition, statistics used to represent extremely short exposures cannot be readily aggregated to provide a representative summary statistic for plant responses resulting from an extended exposure, for example, a growing season (U.S. Environmental Protection Agency 1986). In addition, the exposure statistics currently available were developed chiefly for crop plants, not for forest trees. The absence of reliable exposure statistics makes it extremely difficult to develop a cause-and-effect relationship between air pollutants and forest trees.

To document a cause-and-effect relationship it must be possible to infer a strong pattern of consistency, responsiveness and a proven biological mechanism (Woodman and Cowling, 1987). In simple systems, any two of the above three linkage patterns may be sufficient to infer cause. In more complicated systems, however, all three patterns may be required. Consistency requires that the symptoms of injury and disfunction be consistently associated with the presence of the causal factor; responsiveness the development of

Table 9. Critical Concentrations of Gaseous Pollutants That Induce Visible Foliar Injury to Vegetation in Eastern North America*

Time of Exposure (hr)	SO_2[1]			NO_2[2]			O_3[3]		
	Sensitive ppb	Intermediate ppb	Tolerant ppb	Sensitive ppb	Intermediate ppb	Tolerant ppb	Sensitive ppb	Intermediate ppb	Toleran ppb
0.5	1000-4000	3500-12,000	>10,000	6000-10,000	9000-17,000	>16,000	350-500	550-700	>700
1.0	500-3000	2500-10,000	> 8,000	4000-8000	7000-14,000	>13,000	150-250	350-400	>400
2.0	250-2000	1500- 7,000	> 6,000	3000-7000	6000-12,0000	>11,000	90-150	150-250	>300
4.0	100-1000	500- 5,000	> 4,000	2000-6000	5000-10,000	> 9,000	40- 90	100-150	>250
8.0	50-500	200- 2,500	> 2,000	2000-5000	4000- 9,000	> 8,000	20- 40	70-120	>200

[1]U.S. Environmental Protection Agency. Air Quality Criteria for Sulfur Oxides. Revised Chapter 5: Effects of Sulfur Oxides in the Atmosphere on Vegetation. EPA-R3-73-030. National Environmental Research Center, U.S. Environmental Protection Agency, Research Triangle Park, NC, 1973.

[2]U.S. Environmental Protection Agency. Air Quality Criteria for Nitrogen Oxides. EPA-600/8-82-026. U.S. Environmental Protection Agency, Research Triangle Park, NC, September, 1982b.

[3]U.S. Environmental Protection Agency. Air Quality Criteria for Ozone and Other Photochemical Oxidants. EPA 600/8-84/020OF, Environmental Criteria Assessment Office, Research Triangle Park, NC, August, 1986. Draft Final.

*The majority of the responses cited in the table are based on laboratory experiments.

exposure-response relationships by exposing healthy trees to various known concentrations of airborne pollutants under conditions simulating those found in forests and mechanisms requires that the observed effect be related to the suspected causal factor by one or more biological mechanisms. Responsiveness is the most difficult to prove because exposing large forest trees using controlled conditions is extremely difficult. Extrapolation from exposure using seedlings is not satisfactory either (Pye, 1988).

Sulfur Dioxide.--The adverse effects of SO_2 on vegetation have been observed in cities and around smelters for more than 100 years. The areas in the vicinity of smelters such as those around Sudbury, Ontario, Canada, and Copper Hill, TN, are denuded of vegetation due in large part to SO_2 emissions. Research concerning SO_2 effects on vegetation, for reasons of historical precedent, has been extensively and recently reviewed (U.S. Environmental Protection Agency 1982a; Winner and others 1985;).

Gaseous sulfur dioxide has been rigorously proven to cause visible foliar injury, decreased growth, and mortality of many different species of forest trees in concentrations greater than approximately 50 ppb for 8 hours or longer (see table 9). This knowledge has been accumulated through many years of field studies near strong point sources as well as field and greenhouse experiments (U.S. Environmental Protection Agency 1982a). The majority of the studies dealing with the effects of SO_2 exposure on growth and yield have used crop plants (i.e., annuals). Crop plants have been emphasized because of the possibly greater economic impact if they are injured.

In the eastern United States, the highest sulfur dioxide concentrations have been measured in the Ohio River Valley (see chapter 3, fig. 15). In the southeastern States, exposure to high concentrations is most likely to occur in the vicinity of power plants, metal smelters, and near large cities.

Sulfur is an essential nutrient for plant growth and development. Forest ecosystems require about 4-22 kg/ha/year for normal functions and growth -- mainly synthesis of enzymes, structural and storage proteins, and amino acids. There is a close biochemical relationship between sulfur and nitrogen due to their mutual involvement in amino acid and protein synthesis (Kelly and Lambert 1972). Uptake of sulfur by forest trees, mainly in the form of water soluble sulfate ions, is by absorption through the roots, and through foliar organs from air, cloud water, and precipitation (Turner and Lambert 1980). Thus, low concentrations of gaseous sulfur dioxide (<approximately 40 ppb), and small amounts of airborne sulfate aerosol (up to about 3 kg/ha/year) may enhance growth and uptake of all elements (Turner and Lambert 1980). Atmospheric sulfur additions in polluted areas (10-80 kg/ha/year), however, frequently exceed not only the requirement of the forest ecosystem, but also its ability to accumulate sulfur biologically (Johnson 1984).

Nitrogen Oxides.--Injury to vegetation by NO_x (NO or NO_2) compounds, if it does occur in nature, is presumed to be restricted to the immediate vicinity of industrial point sources (e.g., power plants, arsenals, or nitric acid factories) or to large metropolitan areas. Ambient concentrations of NO_x seldom reach the magnitude (around 2000 ppb) necessary to cause vegetational injury (see table 9). Most vegetational injury associated with NO_x is indirect -- that is, it is the result of photochemical oxidant reactions that produce

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ozone, PAN, and PPN (Kozlowski and Constantinidou 1986a; National Research Council 1977b). Visible injury to forest trees or other forest plants from exposure to concentrations of nitrogen oxides occurs so rarely in eastern North America that there have been no published reports of injury in the field due to NO or NO_2 (National Research Council 1977b; U.S. Environmental Protection Agency 1986).

Nitrogen, like sulfur, is an essential nutrient for plant growth and development. Uptake, mostly as water-soluble nitrate and ammonium ions, is chiefly through the soil. Each year, forest ecosystems in the Southern United States require about 30-75 kg/ha/year of nitrogen for their normal functions and growth -- mainly for synthesis of enzymes, structural proteins, nucleic acids, and many secondary metabolites. These ions, obtained chiefly from soils by absorption through the roots, also can be assimilated through foliar organs from air, cloud water, and precipitation. Approximately a third to a half of the current need for nitrogen (20-60 kg/ha/year) is met by recycling nitrogen through litter-fall. Most of the other half (28-85 kg/ha/year) comes from atmospheric sources and by mineralization of soil nitrogen (Binkley and others 1989).

Concentrations of airborne nitrogen-containing chemicals are being continuously added to forest ecosystems today. The effect of these chemicals on ecosytems has not been clearly determined. Plants and the communities in which they lived were adapted to soils with low nitrogen concentrations and were thriving before anthropogenic nitrogen deposition began. In many areas of North America, current additions of atmospheric nitrogen are in excess of growth requirements, particularly of subalpine and boreal coniferous forests. Only a small fraction of the inorganic nitrogen added to the soil from new sources is absorbed and enhances growth. Most of the nitrogen added is leached from the soil in the form of nitrate (Waring and Schlesinger 1985).

Ammonia (NH_3) and Ammonium (NH_4^+). Ammonia has been known to be a phytotoxic air pollutant since the late 1800's, chiefly because of injury to vegetation in the vicinity of accidental releases of gaseous or liquefied ammonia (National Research Council 1977a; Treshow 1970). Vegetational injury was most commonly associated with release of ammonia from refrigeration systems; however, freons are now being used as heat transfer fluids, so injury from this source has declined.

Agricultural use of anhydrous ammonia and urea as fertilizers is a present-day source of injury to vegetation. In addition, ammonia is added to the atmosphere from a variety of combustion sources, including domestic incineration and automobile engines, as well as volatilization from cattle feedlots and from natural sources (National Research Council 1978b).

Nitrates.--Nitrates are present in the atmosphere as gases (PAN and nitric acid vapor), dissolved in droplets, and also in the form of particulate matter such as ammonium nitrate (NH_4NO_3). Nitrates are secondary pollutants formed in the atmosphere from NO and NO_2 and are an important component of acid deposition (Lovett and Lindberg 1986).

Ozone.--Ozone is the gaseous pollutant most injurious to forests. This conclusion is based upon the massive number of studies involving the responses

of crops to ozone and the somewhat smaller number of studies dealing with tree responses to ozone. All fumigation studies, whether in the laboratory or field, whether on crops or trees, suggest that plants will respond to ozone concentrations above 0.06 ppm within hours (see table 9). Concern stems from the fact that ozone concentrations across most of North America exceed 60 ppb during most of the growing season. Much of our knowledge about plant responses to ozone is summarized in recent volumes (Guderian 1985; U.S. Environmental Protection Agency 1986).

Mixtures of Gaseous Pollutants.--In the vicinity of major pollutant sources such as cities and power plants, the air contains mixtures of gases, aerosols, and coarse particulate matter. Near power plants SO_2 usually is the predominant gas, but NO_2 and O_3 can also be present. The gases may be present simultaneously, sequentially, or intermittently; however, the exposure of forests to more than one of the pollutants simultaneously at concentrations of 30 ppb or higher is unlikely to occur with any appreciable frequency in rural areas farther than 50 km from a point source. The frequency of simultaneous occurrences decreased as the concentration of the gas increased for the months of May through September during the 5-year period, 1978-1982, of the study by Lefohn and his co-workers (1987). Mixtures of SO_2 and NO_2 are experienced most frequently near point sources; mixtures of SO_2 and NO_2 are next, and mixtures of O_3 and NO_2 occur least frequently (Lefohn and others 1987).

Concern regarding pollutant combinations exists because controlled exposure experiments with certain mixtures indicate plants respond to exposures of pollutant mixtures differently from the way they respond to the single pollutant (e.g., mixtures of O_3 and SO_2 together can cause injury at concentrations lower than can either gas by itself). Unfortunately, however, very few of these experiments have included woody plants.

The effects of simultaneous co-occurrences of pollutants (O_3, SO_2, NO_x) on forest trees may not be important if co-occurrences are as rare as reported by Lefohn and Tingey (1984) and Lefohn and others (1987); however, the sequence to which vegetation is exposed to multiple pollutants may be important in causing growth effects.

Mixtures of gases can cause effects that are 1) significantly greater than the additive effects of the gases singly (synergism); 2) significantly less than the additive effects of the gases (antagonism); or 3) not significantly different from the additive effects of the gases acting alone (additive) (Heagle and Johnston 1979).

The likelihood that acid deposition and other airborne sulfur- and nitrogen- derived chemicals will affect the forests of eastern North America depends on the extent that the chemicals enter the trees.

The alteration of physiological processes (photosynthesis, respiration, carbon uptake and allocation), foliar leaching, disruption of regeneration and reproduction, and changes in host susceptibility to pests, pathogens, and mycorrhizal colonization resulting from the exposure of trees to airborne chemical pollutants are discussed in the sections that follow.

4.2.1 By What Physical, Chemical, and Biological Processes Are These Compounds Taken up by Plants, and Microorganisms?

The life-sustaining processes of photosynthesis, respiration, and transpiration involve the exchange of gases between plants and the atmosphere. Exchange of gases during these processes occurs primarily through the stomata on the leaves because the epidermis is coated with a waxy cuticle that is relatively impervious to gases. Exposure to airborne chemical pollutants occurs during the above natural processes.

Uptake of airborne chemical pollutants involves: 1) exposure, i.e., the presence of the chemical pollutant in the vicinity of the forest; 2) deposition into the forest; and 3) movement into the trees through the leaves or from the soil into the roots.

The processes of deposition are the crucial link between transport of atmospheric pollutants to forest canopies, their ultimate uptake and the manifestation of effects on vegetation (Lindberg and McLaughlin 1986). The major problems of how to quantify pollutant dose so that both chemical species and exposure kinetics are identified, and how to characterize plant response so that actual or potential economic or ecologic losses can be quantified, are related to gas exchange at the atmosphere-leaf interface (Lindberg and McLaughlin 1986). The gas-exchange processes for the common gaseous species such as O_3, SO_2, HNO_3, and NO_2 have been parameterized in dry deposition models (Baldocchi and others 1987).

Differences in the rates of pollutant deposition could, in part, account for the variation in plant responses that are usually considered a function of genotype, environment, pollutant exposure dynamics (movement of pollutants into the leaves), or the presence of other pollutants. Because the physiological sites of action for most gaseous pollutants are cells in the leaf interior (table 10), any factor that influences pollutant deposition (e.g., rate, concentration, form) will change the relationship between the concentrations of the pollutant in the atmosphere and the corresponding concentration at the sensitive sites within leaf cells. An understanding of the role of deposition processes should, therefore, aid in explaining patterns in pollutant effects on the physiological status, growth, development, and productivity of plants (Taylor and others 1988).

Table 10. Types of Interactions of Gases With Plant Surfaces

Adsorb to or react with all surfaces	H_2O_2, HF, HCl, HNO_3
Interact primarily inside leaf tissue	SO_2, NH_3, Cl_2, NO_2, H_2CO, O_3, PAN, H_2O*, metabolically reactive CO_2*, O_2*
Exchange slowly with plants; potentially important interactions	N_2O, NO, CO, organic molecules such as hydrocarbons, pheromones, ethylene, and terpenes

*Normal large-scale exchange in metabolic processes.

Source: Hosker and Lindberg (1982).

A variety of physical, chemical and biological factors control deposition rates (Bennett and Hill 1975). These factors include the micrometeorological conditions above and within the plant canopy, leaf surface characteristics (shape, degree of pubescence, and surface chemistry) and the chemical and biological properties of individual pollutants, such as diffusivity, water solubility, and biological reactivity within cell systems (Lindberg and McLaughlin 1986).

Pollutant deposition can be addressed from different levels of organization: 1) at the plant canopy level, focusing on atmospheric processes governing turbulence in terrestrial landscapes and the distribution of pollutant deposition sites within plant canopies; 2) at the level of the individual leaf, focusing on foliar sites of deposition and the role of cuticular, stomatal, and mesophyll resistances in governing trace gas rates of exchange (Bennett and others 1973); and 3) at the molecular level, emphasizing the physiochemical processes at the intercellular space and mesophyll cell surface interface in the leaf interior (Tingey and Taylor 1982);

Dry deposition of gaseous pollutants to plant canopies and individual leaves occurs because a chemical gradient exists between the atmosphere and the deposition sites, either on the exterior leaf surfaces or on cells of the leaf interior. Dry deposition involves both gases and particles. Deposition of particles and cloud water to canopies and individual leaves, however, is governed primarily by processes other than diffusion. Vertical turbulence transports particles across the atmospheric boundary layer to the laminar boundary layer of individual leaves. Atmospheric turbulence, which is governed by mechanical mixing, buoyancy, and surface roughness, increases deposition. Particle cloud/water deposition is governed by gravitational settling, droplet impaction, interception efficiencies, and Brownian diffusion (Taylor and others 1988).

During wet deposition gases and particles are delivered to the canopy partly in solution, thus enhancing the possibility of sorption by foliage. Other forms of wet deposition such as dew, mist and/or fog interception can create conditions on the leaf surface that lead to extremely high concentrations of pollutants because of interactions with deposited particles and gases (Hosker and Lindberg 1982). Wet deposition is episodic and delivers the pollutants in highly irregular, intense concentrations. In addition to depositing pollutants on plant surfaces, wetfall may simultaneously remove previously deposited substances, thus redistributing pollutants. Therefore, the net effects of precipitation may be either to increase or decrease the surface concentration of a particular pollutant (Lindberg and others 1979).

Surface exchange processes occurring between the atmosphere and plant canopies present special problems in mass transport of air pollutants. Fluxes of air pollutants and materials such as CO_2 and H_2O within the canopy vary because the vertical distribution of sources and sinks also varies with canopy height, the species composition, and with the physiological state of plants. Gases react with the various types of surfaces found within the vegetative canopies, especially leaves. Leaf surfaces of different species vary in degree of pubescence, number of trichomes, and in the presence of wax and exudates. The effectiveness of these competing sinks is not equivalent. The interaction

of gases with these leaf components may alter the net effect of any individual gas on vegetation (Heagle and Johnston, 1979; Hosker and Lindberg 1982; Taylor and others 1988).

When gases or particles approach plant leaves, they must eventually pass through the inner portion of the leaf boundary layer. Transfer within this area is no longer governed by turbulence effects. The leaf-air microenvironment within this sublayer may differ substantially from that outside the boundary layer. Gases moving into the leaf encounter concentrations of transpired water vapor and a variety of hydrocarbons such as terpenoids that are produced in secondary plant metabolism, volatilized into the gas phase of the leaf interior, and emitted by the leaf through the stomata. These substances are higher within the inner boundary layer and may significantly affect the surface uptake of gases and particles (Hosker and Lindberg 1982).

Gas exchange at the atmosphere-leaf interface has certain unique aspects. These are: (1) a pronounced and viscous/stagnant layer in the gas phase adjacent to the leaf surface (boundary layer) and in the leaf interior (sub-stomatal chamber and intercellular space); (2) a variable diffusive resistance in the gas phase due to changes in stomatal porosity; (3) a distributed array of extensive deposition sites both on the leaf surface and within the leaf interior; (4) a biochemically variable capacity for trace gas deposition and assimilation in the mesophyll tissues; and (5) at the level of the plant canopy, a mix of functionally varying leaf surfaces comprised of different age classes and species, each having its own distinct local microclimate such that the canopy's effective deposition/sink potential varies in space and time.

Stomatal control of diffusion into the leaf is a major factor influencing the exchange of gaseous pollutants between the atmosphere and the internal tissues of leaves. All studies indicate that stomatal resistance (the extent to which stomata are open or closed) is the most dynamic and most influential resistance to O_3, SO_2, and NO_2 transfer when the canopy is dry (Baldocchi and others 1987). Functioning of stomata in turn is controlled by both physiological and bioenvironmental processes (Hosker and Lindberg 1982; Wesely and Hicks 1977; Winner and others 1985). The rate of diffusion through the stomata depends on (1) the diffusivity of the gases, (2) the magnitude of the concentration gradient for a specific gas, and (3) the degree to which stomata are open (Winner and others 1985).

The transfer of gases from the atmosphere to the leaf interior involves both a gas phase and a liquid phase. The leaf interior is a highly porous and physiologically complex organ. Given the leaf's morphology, the ease of diffusion and site deposition for gaseous pollutants are likely to vary appreciably among gases in accordance with their physical and chemical properties (Taylor and others 1988).

The gas-phase flux depends upon the concentration gradient between ambient air and the wet cell surfaces within the leaf and the resistance to mass transfer along the diffusion pathway. Within the leaf, following deposition on wet cell surfaces, flux is governed by liquid-phase processes including diffusion, formation of pollutant derivatives, and productivity of plants (Tingey and Taylor 1982).

Uptake of some dry deposited gaseous pollutants (sulfur dioxide, nitrogen oxides, ozone, peroxyacyl nitrates, nitric acid, ammonia, and hydrogen peroxide) occurs by absorption and adsorption of the gas on the moist walls of mesophyll cells. After diffusing into the intercellular space of the leaf interior, gas molecules are partitioned across this gas-to-liquid interface on cell surfaces at a rate determined by the solubility of the gas in the external cell solution and the chemical reactivity of the gas in aqueous phase (Taylor and others 1983). Uptake of NO, particularly, is controlled by its extremely low solubility (Malhoutra and Kahn 1984). Tingey and Taylor (1982) have proposed that deposition sites for highly reactive water-soluble gases such as HNO_3 and SO_2 are the leaf surface and the substomatal cavity, whereas, the site for less soluble gases (e.g., O_3) is the mesophyll tissue (Taylor and others 1988).

Uptake of sulfate, nitrate, ammonium, and hydrogen ions and nutrient cations and anions in fine and coarse particles, in coarse particulate matter, and in precipitation and cloud water after impaction or gravitational settling on vegetation surfaces, occurs more slowly than uptake of gases. Uptake of these ions after dissolution occurs mainly by diffusion through the cuticle on leaves or through the bark, and by diffusion through fine feeder roots and mycorrhizae after percolation and diffusion through the soil.

Absorption rates depend on pollutant concentration, cuticle thickness and hydration, and the affinity of cuticular compounds for the solutes involved. Soluble compounds may enter leaves through cuticular breaks, trichomes, wounds, or stomata. Leaf exudates also may play a role in the entrance of deposited substances (Hosker and Lindberg 1982).

Lichens and mosses, important nonvascular plants growing in forests, have low tolerances to air pollutants (Winner and Bewley 1978a, 1978b). Their sensitivity is greater, in part, because these plants accumulate airborne compounds at a faster rate than vascular plants do (Winner and others 1978). Lichens are composed of a fungus and an alga. The thallus (plant body) of lichens, unlike the leaves of vascular plants, have no waxy cuticle. Bare patches, holes, pores, and depressions on the surfaces of the lichen permit the interior of the thallus to be exposed to ambient air all times of the day or night. For this reason, airborne chemicals can diffuse rapidly into the thallus where they can accumulate and cause injury. The algal cells of lichens are generally more sensitive to injury by sulfur dioxide and ozone than the fungal cells (Anderson and Treshow 1984).

Mosses have low tolerances to air pollutants. Their leaves are usually a single cell thick and have no cuticle, therefore, readily absorb airborne chemicals. Winner and others (1978), by using sulfur isotopes, showed that mosses absorb airborne sulfur. Because they can accumulate large quantities of sulfur rapidly, they are more vulnerable to effluents such as SO_2 than vascular plants (Winner and others 1978).

Exposure of plants to airborne gases fine or coarse aerosols, or coarse particulate matter through the soil, is indirect. Four separate processes are involved:

- Deposition;
- Dissolution into the soil water;
- Percolation or diffusion through the soil; and finally,
- Absorption through the surfaces of fine feeder roots or mycorrhizae.

Many biotic, physical, and chemical factors influence these processes. The emphasis of this section is on foliage-mediated effects; therefore, the soil-mediated exposures are not discussed at this time.

Microorganisms readily absorb sulfur- and nitrogen-containing compounds from soil in forest ecosystems. Microorganisms can immobilize nutrients and reduce the quantity in the soil available to plants via nutrient assimilation (Alexander 1977).

In summary, the physical, chemical, and biological processes involved in plant uptake of sulfur- and nitrogen-derived compounds from both air and soil are influenced by a variety of biotic and abiotic factors. The biotic factors include

- stage of plant development;
- genetic variation in plant species;
- competition among several species and individuals of the plant community for light, water, growing space; and
- interaction among the pollutant, the plant, plant pathogens and pests, and root mycorrhizal fungi.

The abiotic factors include

- photoperiod;
- light intensity;
- temperature;
- relative humidity;
- amount of rainfall and its timing;
- soil moisture;
- soil fertility;
- interactions among co-occurring pollutant chemicals; and
- dewfall (Heck 1982).

4.2.2 What is the Mode of Action of the Different Airborne Chemical Pollutants?

Gaseous pollutants must enter plants to cause an effect. Alteration of physiological processes begins at the biochemical level within the cells of leaves. Entrance is mainly through the stomata in the leaves, though some entrance may occur through the cuticle, especially if it is weathered or damaged. Pollutant uptake is controlled by the stomata and the environmental conditions that determine stomatal behavior. Any action of a pollutant on stomatal behavior affects gas exchange and water loss from plants (Black 1985). Leaf (i.e., stomatal) conductance regulates the movement of the gas from the leaf boundary layer into the leaves (Black 1985; Tingey and Taylor 1982; Winner and Mooney 1980a, 1980b). The mode of action of sulfur- and nitrogen-containing gaseous pollutants, of ozone, and of mixtures of these pollutants is discussed in the sections that follow.

Sulfur dioxide, nitrogen oxides, ozone and PAN (and possibly hydrogen peroxide, H_2O_2), the major pollutants discussed in this chapter, have a number of features in common:

1) All five pollutants are gases that enter plants primarily through stomata;

2) At least the first three alter the normal regulatory function of guard cells and thus can increase or (usually) decrease stomatal conductance;

3) All five are highly reactive oxidizing agents, and at least the first three attack the membranes and organelles of cells and thus alter general metabolic processes;

4) The first four are known to inhibit photosynthesis, increase respiration, and alter carbon allocation between shoots and roots.

a) Sulfur Compounds

Sulfur Dioxide.--Uptake of SO_2 through stomata occurs mainly in the summer. Stomatal activity in winter is low, although uptake in even subzero temperatures may occur in some evergreen trees. Deciduous trees also can absorb SO_2 in winter if their lenticels are active (Garsed 1985).

There is no simple pattern of stomatal response to SO_2. Exposure to SO_2 may induce either increased stomatal opening or closure in the same species. Low concentrations in some studies increased stomatal opening, while high concentrations induced closure (Black 1985). Responses to SO_2 are so diverse and depend on so many factors, it is extremely difficult to assess the long-term implications of exposure. Within a forest canopy, stomatal responses vary spatially and with time in concert with variation in pollutant concentrations, environmental conditions, and the biological state of each particular plant part (Black 1985).

Regardless of the ambient air concentration of SO_2 to which trees are exposed, tree response is determined by the amount of SO_2 that enters the leaves of the trees. After diffusing through the stomata, SO_2 dissolves in the water on the cellular surfaces to form sulfite (SO_3^{2-}), bisulfite (HSO_3^-) and, depending on the pH of the surrounding medium, other ionic species (Malhoutra and Kahn 1984). Both sulfite and bisulfite are toxic to many biochemical and physiological processes (Malhoutra and Hocking 1976; Ziegler 1975).

Plants can detoxify sulfite and bisulfite by further oxidizing them to sulfate. Toxicity is determined by the site at which conversion occurs. Toxicity may occur if, during short episodes with high SO_2 concentrations, the rate of SO_2 conversion to sulfate is exceeded (Ziegler 1975). At low SO_2 concentrations, plants can accumulate sulfur compounds; however, chronic symptoms may appear if the capacity of a plant to metabolically incorporate sulfate is exceeded. Detrimental toxic effects are usually associated with high SO_2 concentrations, while growth stimulation or no effects may be associated with exposure to low concentrations (U.S. Environmental Protection Agency 1982a).

Sulfate Aerosols.--Sulfate aerosols are secondary pollutants formed in the atmosphere through the oxidation of SO_2 to sulfates. Sulfates (SO_4^{2-}) are an important component of acid deposition, but studies dealing with the effects of sulfates are limited. The majority of the studies reporting the effects on vegetation of sulfate-containing acid deposition emphasize the hydrogen ion, the associated changes in pH, and the effects of the gaseous components (Altshuller and Linthurst 1984).

Aerosols must be soluble to enter leaves through stomata, trichomes, or cuticular breaks and wounds before injury can occur at the cellular level within a plant (Hosker and Lindberg 1982). Chevone and others (1986) suggest that sulfuric acid aerosols can enter leaves through the stomata; however, no visible injury was noted in soybeans exposed to acid aerosols (500 $\mu g/m^3$ for 4 hours), a concentration much greater than is usually observed in the ambient air. Foliar injury was observed, however, when pinto beans (Phaseolus vulgaris L.) were exposed for a 3-week period to submicrometer ammonium sulfate aerosols using an aerosol concentration two orders of magnitude above the ambient (Gmur and others 1983). Leaf injury was probably due to the ammonium fraction of the aerosol as sulfate in leaves has not been shown to be injurious (Nihlgard 1985). No field studies dealing with the uptake of sulfate have been found in the literature (Krupa and Legge 1986).

b) Nitrogen Compounds

Nitrogen Oxides.--Gaseous nitrogen compounds, principally NO and NO_2, must diffuse through stomata or cuticle to cause a plant effect. Perturbation within mesophyll cells in the leaf results after gaseous NO or NO_2 from the ambient air enters the stomata, dissolves in the extracellular water on the moist cell surfaces, and reacts to form nitrous oxide which then dissociates to nitrite (NO_2^-). The solubility of NO and NO_2 in the extracellular water is a major influence in determining the rate of uptake. While NO is poorly soluble, NO_2 is highly soluble (Mansfield and Freer-Smith 1981). The solubility of NO in xylem sap is greater than in distilled water. This fact may play an

important role in its uptake because xylem sap is continuous with the extracellular water in a leaf (Anderson and Mansfield 1979). Both stomatal and mesophyll resistance control uptake (Srivastava and others 1975a).

Plants have the ability to metabolize the dissolved NO_x via the following general metabolic pathway: $NO_x \rightarrow NO_3^- \rightarrow NO_2^- \rightarrow NH_4^+ \rightarrow$amino acids$\rightarrow$proteins. Nitrite is usually considered to be a toxic ion, though not all scientists agree. Available evidence suggests that effects of NO_x are a consequence of increased nitrite and nitrate concentrations within plant cells, particularly nitrite. Normal nitrogen metabolism follows the pathway indicated above. If amino acids are not formed, toxic compounds will accumulate. Injury results when nitrite reduction in leaves is inhibited (Mansfield and Freer-Smith 1981). Plants that have been exposed to NO_2 exhibit increased concentrations of nitrate reductase (NiR) in the leaves. Plants exposed to both NO_2 and SO_2 failed to show an increase in nitrate reductase in the leaves (Mansfield and Freer-Smith 1981). Most of the effects of NO_x on nitrate metabolism can be explained by assuming pollutant-induced changes in the rates of reduction of NO_3^- and NO_2^- ions by plants (Rowland and others 1985).

Ammonia (NH_3) and Ammonium (NH_4^+).--The concentrations required to cause visible vegetational injury have not been well quantified. Foliage is the most susceptible plant part (National Research Council 1977a). Ammonia dissolves in the waterfilm on the walls of the mesophyll cells after entrance through the stomata. This view is supported by experiments in which pinto bean plants were exposed for 12 to 21 days to submicrometer ammonium sulfate aerosols at concentrations two orders of magnitude above the ambient concentration of 60-100 $\mu g/m^3$. Visible injury was observed along the leaf margins; however, initial injury was to the spongy mesophyll cells (Gmur and others 1983). Experiments using $^{15}NH_3$ indicate that once inside the cell, ^{15}N is incorporated into amino acids and proteins and ultimately is transported to the roots (van Hove and others 1987).

Nitrates.--Dry deposition of nitrates into forests can be an important source of inorganic nitrogen. Lovett and Lindberg (1986) reported that dry deposition of atmospheric nitrates supplied nearly 60 percent of the inorganic nitrogen deposited in the Walker Branch Watershed in eastern Tennessee. Canopy net uptake was estimated to have been 3.2 kg NO_3^--N/ha/year. Canopy uptake of deposited nitrate is considered to be a general phenomenon in forest ecosystems. Taylor and others (1988) point out that the deposition site for HNO_3 vapor is the leaf surface and the substomatal cavity. Nitrates, as well as sulfates deposited on the soil, can enhance growth. The fate of nitrate aerosols after deposition on plant foliage is an area needing study.

Peroxyacetyl Nitrates. PAN is the most common member of a series of homologues that increase in phytotoxicity with increase in molecular weight. Only PAN is found in ambient air at concentrations that are injurious to vegetation, and then only in limited areas of the country (U.S. Environmental Protection Agency 1986). Because concentrations injurious to vegetation have not been reported from the eastern United States, and because woody plants are not very susceptible to injury by PAN, its mode of action will not be discussed here.

Only ozone that enters the plant through the leaf stomata can impair plant processes. An effect will occur only if sufficient ozone reaches the sensitive sites within a leaf. The effects range from subtle modifications of cellular biochemistry and whole plant physiology to visible injury.

c) Ozone and Hydrogen Peroxide

Ozone.--Ozone enters the leaf through stomata; once within the leaf it quickly dissolves in the aqueous layer on the cells lining the air spaces. Ozone, or its decomposition products, then diffuses through the cell wall and membrane into the cell, where it may affect cellular metabolism. At any point along this pathway, ozone or its decomposition products may react with cellular components. Altered cell structure and function may result in changes in membrane permeability, carbon dioxide fixation, and many secondary metabolic processes (Tingey and Taylor 1982).

Ozone injury will not be detected if (1) the rate of ozone uptake is sufficiently small so that the plant is able to detoxify or metabolize ozone or its derivatives; or (2) the plant is able to repair or compensate for the ozone impacts (Tingey and Taylor 1982).

The uptake and movement of ozone to sensitive cellular sites are subject to various physiological and biochemical controls. The magnitude of ozone-induced effects will depend upon the physical environment of the plant, including macro- and microclimatic factors; the chemical environment of the plant, including other gaseous air pollutants; and biological factors, including genetic potential, developmental age of the plant, and interaction with plant diseases and insects. Cellular injury may subsequently manifest itself in a number of ways, including visible foliar injury; premature senescence; reduced growth or yield, or both; reduced plant vigor; and sometimes death.

Some scientists consider membranes to be the primary site of action of ozone (Heath 1980; Tingey and Taylor 1982). Alteration in plasma membrane function is an early event in the sequence of ozone-induced effects that eventually leads to leaf injury and subsequent growth reduction. Changes in the semipermeability of the membrane are evidenced by changes in fluxes of carbohydrates, amino acids, inorganic ions, and water (Heath 1975, 1980; Tingey and Taylor 1982). These functional changes lead to cellular dysfunction and death similar to that induced by water stress and pathogenesis (Heath 1975; Thomson 1975; Tingey and others 1976b).

Hydrogen Peroxide (H_2O_2).--Study of plant response to H_2O_2 is in the early stages. Mallant and others (1986) reported injury to Norway spruce (Picea abies) based on experimental exposures to an acidic mist containing H_2O_2. They reported a significant decrease of cell area in the mesophyll in both primary and secondary needles and also in the vascular bundles; however, in some cases changes were found only in the primary needles. Movement into the leaf was considered to be through the stomata. The authors state that H_2O_2 must be considered an important factor in forest dieback in Europe. At present this view is subject to question, as scientists in the United States have not been able to duplicate the work of Mallant and others (1986).

d) Mixtures of Gaseous Pollutants

Little is known about how stomatal regulation and associated biochemical and physiological processes are influenced by mixtures of pollutants (Malhoutra and Kahn 1984). The effects of pollutant mixtures are complex and differ with species, cultivar, and experimental conditions (Ormrod 1982). The effects of mixtures of gases on plants are possibly mediated by the following:

1) reactions between the pollutant gases after entering the leaf; 2) an effect by one of the gases on the stomatal aperture; 3) competition between the gases for the reaction sites; 4) a change at the reaction sites in sensitivity; or 5) a combination of these effects (Heagle and Johnston 1979).

Mixtures of Gases and Aerosols.--Fine particulate aerosols and ozone are formed concurrently in the atmosphere. The interaction of particulate sulfate aerosols and ozone is a possible reason for concern. Fine particulate aerosols accumulate at night in a reactive plume, while ozone decreases at night and then peaks during the daytime hours. Thus, because of the diurnal photochemical patterns of ozone, vegetation is first exposed to peak sulfate aerosols concentrations before exposure to ozone (Krupa and Legge 1986). Under these circumstances when conditions are acidic, the residence time of ozone is significantly increased (Chevone and others 1986; Krupa and Legge 1986).

During periods of wet deposition, ambient ozone concentrations are generally below detection limits; however, the acidification of receptor surfaces could predispose foliar surfaces to subsequent ozone exposure. Laboratory studies suggest this possibility. Neither soybean (Glycine max L.) nor pinto bean (Phaseolus vulgaris L.) exhibited visible foliar injury when exposed to sulfuric acid aerosols (500 $\mu g/m^3$ for 4 hours); however, when exposed to ozone at 190 ppb for 4 hours, visible injury was observed (Krupa and Legge 1986). Chevone and others (1986) reported similar results using similar concentrations of ozone and acid aerosols. Additional studies are needed to determine whether predisposition occurs when sulfuric acid aerosols at concentrations near ambient are used in the exposures.

e) Acidic Deposition

Acidic deposition, in wet form, is a dilute solution of hydrogen ions and associated anions. This solution, depending on the surface it comes into contact with, is easily altered. If the surface is basic, the pH increases; if acidic, the pH may decrease (Bennett and others 1985). The soluble ions usually encountered in precipitation are hydrogen (H^+), bicarbonate (HCO_3^-), calcium (Ca^+), magnesium (Mg^+), sodium (Na^+), potassium (K^+), sulfate (SO_4^-), nitrate (NO_3^-), chloride (Cl^-), and ammonium (NH_4^+). Sulfate and nitrate ions are the dominant ones in precipitation in the East (Stensland and others 1986).

Much has been written about the possible effects of acidic deposition on forests and its relationship to forest decline. No evidence to date supports the concept that there is a direct relationship between acidic deposition and forest effects. A possible indirect effect of acidic deposition on forests via soil-mediated response mechanisms has been hypothesized (Amthor 1984; Bennett and others 1985; National Acid Precipitation Assessment Program 1987;). In a separate document, Binkley and others (1989) discuss soil-mediated responses.

Summary.--Gaseous atmospheric pollutants must enter plants, chiefly through the stomata, to cause a response. Pollutant chemicals in the gas phase diffuse through the stomata into the substomatal cavity where they enter the liquid phase by dissolving in the water film on the walls of the cells lining the substomatal cavity. From this point along this pathway, the phenomena that influence the behavior of the pollutant chemicals are not well understood. The cell membrane has been suggested as the site of ozone-induced perturbation

(Tingey and Taylor 1982). Other pollutants may also move through the membrane into the cell. Perturbation, changes in cell structive or function, may result. The physiological and biochemical changes that cause perturbation are not well understood for the majority of the pollutants. Sulfur dioxide, nitrogen oxides, and ozone have been studied the most. Response of plants to nitrogen oxides is based solely on experiments conducted in the laboratory, greenhouse, or in the field under simulated ambient conditions. At present no data exist indicating that vegetation in the field in the United States has been injured by nitrogen oxides alone.

Acidic deposition has not been shown to have an adverse effect on forests via foliage-mediated response mechanisms.

4.2.3 Is Photosynthesis in Forest Trees Altered by Exposure to Airborne Chemical Pollutants?

Movement of pollutant chemicals through stomata into cells in the leaf interior can alter cellular function. Inhibition of photosynthesis is one of the physiological changes attributed to the entrance of airborne pollutant chemicals into plant leaves.

Carbon is the basic building block of the large organic molecules necessary for life. Plants accumulate, store, and use carbon to build their structure and maintain their physiological processes. Plants obtain carbon from the atmosphere and during the process of photosynthesis use sunlight to convert carbon dioxide (CO_2) into carbohydrates. Carbohydrates serve as the raw materials for further biochemical synthesis. Photosynthesis takes place in chloroplasts that are located in the palisade parenchyma and spongy mesophyll cells of leaves (Waring and Schlesinger 1985).

From the atmosphere, gaseous carbon dioxide diffuses through the stomata into the substomata cavity and the intercellular spaces of the leaf. Then, in the liquid phase, it diffuses through the cell membrane into the palisade and spongy mesophyll cells (Waring and Schlesinger, 1985). Alteration or disruption of the movement of carbon dioxide into tree leaves can affect the process of photosynthesis.

a) Sulfur Compounds

Sulfur Dioxide. Inhibition of photosynthesis often is regarded as the first sign of SO_2 toxicity in plants (Darrall 1986; Hällgren 1978). Pollutants, such as SO_2, have a direct effect on the photosynthetic capacity of leaves even at very low concentrations. The rate of photosynthesis in a leaf at any time is governed by certain physiological factors, including stomatal conductance; the nutrient content of leaves; and the biochemical integrity of organelles, membranes, and enzymes in the leaf mesophyll cells (Winner and others 1985). By altering stomatal conductance or by changing the metabolic capacity of mesophyll cells, SO_2 can affect photosynthesis (Winner and Mooney 1980a; Ziegler 1975).

Sulfur dioxide, regardless of whether it induces stomatal opening or closure, generally has a negative influence on photosynthesis (Winner and others 1985). At least initially, decreased photosynthesis is related to decreased stomatal conductance. Leaves with the highest conductance, (the least stomatal resistance to the entry of atmospheric gases) will receive the

largest amount of pollutants in the mesophyll per unit of time. Any predictor of photosynthetic capacity is also a predictor of stomatal conductance, and hence of the potential sensitivity of leaves to injury from SO_2 and other pollutants as well (Winner and others 1985).

Photosynthesis is one of the first processes affected by short-term SO_2 exposures. A 10 percent reduction in photosynthesis was observed within 30 minutes after exposure of Vicia faba L. to SO_2 at 70 ppm (180 $\mu g/m^3$) (Black and Unsworth 1979). Inhibition of net photosynthesis, on the other hand, was observed within 10 minutes when two varieties of Vicia faba L. and four other plant species were exposed to high SO_2 concentrations of 1020 ppb (2,662 $\mu g/m^3$) (Darrall 1986). Recovery commenced immediately upon cessation of fumigation. Darrall (1986) further observed that the rate of recovery was dependent on the level of inhibition at the end of the experiment. Recovery was complete within 2 hours if inhibition of photosynthesis did not exceed 20 percent. Little residual effect was observed from a single exposure if the exposures were 24 hours apart; however, exposures at more frequent intervals where recovery was incomplete, decreased the rate of net photosynthesis in each subsequent recovery period. Recovery of photosynthesis after termination of fumigation was also observed by Black and Unsworth (1979), Bennett and Hill (1973), and Sij and Swanson (1974).

Inhibition of photosynthesis in saplings of woody plants has also been observed. Carlson (1979) exposed native saplings of sugar maple (Acer saccharum Marsh), black oak (Quercus velutina Lam.), and white ash (Fraxinus americana L.) to continuous exposures of 500 ppb (1,310 $\mu g/m^3$) of SO_2 for either 1 to 2 days, for 1 week, or to intermittent exposures of 4 hours per day for 3 weeks (table 11). Photosynthesis was reduced at all exposures, but for each species the percentage reduction was different. The decrease occurred without visible injury. Keller (1977) also reported a reduction in the rate of photosynthesis when grafted 3-year-old potted silver fir (Abies alba Miller), Norway spruce [Picea abies (L.) Karst.], and Scotch pine (Pinus sylvestris L.) were exposed to three concentrations of SO_2, 50, 100 and 200 ppb (130, 260, 520 $\mu g/m^3$), respectively, for 10 weeks each during the spring, summer, and fall, respectively. The rate of photosynthesis decreased as concentration and duration of fumigation increased. No visible injury symptoms were observed.

Reduction in the photosynthetic capacity of trees, if continued over time, could decrease growth and weaken trees, causing them to be more susceptible to insect and pathogen attack, as well as other stresses. Concentrations such as those used in the majority of studies cited above are seldom encountered in the ambient environment except around power plants and metal smelters.

b) Nitrogen Compounds

Under experimental conditions in the laboratory, both NO and NO_2 inhibit apparent photosynthesis in herbaceous plants at concentrations and exposure periods that do not cause visible injury. The magnitude of inhibition of photosynthesis is reported to increase with NO_2 concentration and duration of exposure (Hill and Bennett 1970; Srivastava and others 1975a). Inhibitory action was greatest during the first hour of exposure and then began a continuous decline at a slow rate as long as the exposure continued (Hill and Bennett 1970; Srivastava and others 1975a). Inhibition of photosynthesis was not

Table 11. Effects of SO_2 on Photosynthesis in Tree Saplings

Plant Species	Type of Exposure	Time	Concentration ppm	Concentration $\mu g/m^3$	Effects*
Sugar Maple	C	1-2 days	0.5	1310	22% reduction in photosynthesis
Sugar Maple	C	24 hr/day for 1 wk	0.5	1310	54% reduction in photosynthesis
Sugar Maple	I	4 hr/day for 3 wks	0.5	1310	43% reduction in photosynthesis
Black Oak	C	1-2 days	0.5	1310	40% reduction in photosynthesis
Black Oak	C	24 hr/day for 1 wk	0.5	1310	48% reduction in photosynthesis
Black Oak	I	4 hr/day for 3 wks	0.5	1310	43% reduction in photosynthesis
White Ash	C	1-2 days	0.5	1310	27% reduction in photosynthesis
White Ash	C	24 hr/day for 1 wk	0.5	1310	20% reduction in photosynthesis
White Ash	I	4 hr/day for 3 wks	0.5	1310	7% reduction in photosynthesis

C and I indicate continuous and intermittent exposure.

*No visible symptoms accompanied reduction in photosynthesis.

Source: Carlson (1979).

permanent. Recovery began immediately with the termination of exposure. The time required for complete recovery depended on the magnitude of inhibition (Hill and Bennett 1970).

The concentrations of NO_2, PAN, and PPN and the period of exposure required to inhibit photosynthesis differ with the plant species or cultivar, leaf age, humidity, temperature and amount of light (Srivastava and others 1975b; U.S. Environmental Protection Agency 1986). Nitric oxide (NO) was reported to be approximately half as effective in reducing apparent photosynthesis in alfalfa and oats as NO_2; however, the decrease in the rate of photosynthesis as well as the rate of recovery was more rapid (Hill and Bennett 1970). Photosynthesis in beans was significantly depressed by nitrogen dioxide concentrations of 1,000 ppb or higher for exposures of half an hour or longer. The degree of inhibition was increased by increasing NO_2 concentration and increasing exposure time (Srivastava and others 1975a). The percentage of inhibition changed with leaf age and was highest for leaves that had the highest rates of photosynthesis. The reduction of photosynthesis to a greater extent than transpiration or stomatal conductance suggests that the inhibitory effect of NO_2 occurred within the leaf and not at the point of CO_2 entry.

The effects observed in the studies discussed above occurred at exposures that have never been reported (1,000 ppb for 1 hour or longer) in the ambient air with the possible exception of the highest concentrations observed around a very few strong emission sources. Capron and Mansfield (1976), however, observed a reduction in the photosynthetic rate of tomato (Lycopersicon exculentum Mill.) plants exposed to 250 ppb of NO_2 and 250 ppb of NO or higher concentrations over a 20-hour period. The effect of the two gases on combination resulted in an additive inhibition of photosynthesis.

c) Ozone

The effects of O_3 on leaf gas exchange are not as well known as for SO_2. This is surprising, since O_3 is considered to be the more injurious pollutant of the two. However, few systems have been built which permit the introduction of O_3 into leaf cuvettes so that dynamic leaf responses to O_3 can be measured. Lack of such systems also prevents studies that define the effects of O_3 on the light-harvesting efficiency of leaves, the activity of RuBP carboxylase, and other diagnostic features necessary for assessing mechanisms of O_3 injury. Consequently most research using O_3 involves fumigating plants in one chamber, then turning off ozone or moving the plants to another site before measuring photosynthesis and other gas exchange parameters.

Ozone has the potential for inhibiting photosynthesis. Species in which ozone has been shown to reduce photosynthesis are northern red oak (Quercus rubra L.) (Reich and Amundson 1985), loblolly pine (Pinus taeda L.), slash pine (P. elliottii Engelm. ex Vasey) (Barnes 1972a), ponderosa pine (P. ponderosa Dougl.x Laws) (Coyne and Bingham 1981; Miller and others 1969), eastern white pine (P. strobus L.) (Barnes 1972a; Reich and Amundson, 1985; Botkin and others 1971, 1972), black oak (Quercus velutina Lam.), sugar maple (Acer saccharum Marsh.) (Carlson 1979; Reich and Amundson 1985), and one poplar hybrid (Populus deltoides x trichocarpa) (Reich and Amundson 1985) (see table 12).

Table 12. Effect of Ozone on Photosynthesis

Species	O_3 Concentration ppm[a]	Exposure Duration	% Inhibition	Reference
Loblolly pine	0.05	18 wk continuously	15[b]	Barnes (1972a)
Slash pine	0.05	18 wk continuously	9[b]	Barnes (1972a)
Ponderosa pine	0.15	9 hr daily/60 days	25[c]	Miller and others (1969)
	0.30	9 hr daily/30 days	67[c]	
Eastern white pine	0.15	19 days	10[c]	Barnes (1972a)
Eastern white pine	0.10	4 hr/daily for 50 days	24[b]	Yang and others (1983)
Sensitive	0.20	4 hr/daily for 50 days	42[b]	
	0.30	4 hr/daily for 50 days	51[b]	
Intermediate	0.10	4 hr/daily for 50 days	Not sig. diff.	
	0.20	4 hr/daily for 50 days	14[b]	
	0.30	4 hr/daily for 50 days	20[b]	

[a] 1 ppm = 100 ppb

[b] $P < 0.05$

[c] $P < 0.01$

Source: Adapted from U.S. Environmental Protection Agency (1986).

Some of the studies cited above involve ozone fumigation experiments with concentrations at or below 120 ppb; these are of particular value because they occur in the ambient atmosphere. Barnes (1972a) examined the impact of ozone on seedlings of three species of pine at concentrations of 50 or 150 ppb continuously for 19 days to 18 weeks. In younger seedlings of eastern white pine, which bore only primary needles, ozone had little influence on rate of photosynthesis. In older seedlings with secondary needles, photosynthesis was slightly depressed. With seedlings of slash, eastern white, and loblolly pines, exposure at 105-ppb ozone had a relatively consistent depressing influence on photosynthesis of all species. At 50 ppb, however, ozone appeared to stimulate photosynthesis in older secondary needles and to depress photosynthesis in younger secondary needles. Barnes (1972a, 1972b) used a Mast meter to measure ozone; the Mast meter can underestimate the ozone concentration unless it is calibrated against a reference standard. Also, the sample size used in these experiments was very small, four to nine seedlings. It is possible that variation among samples may have masked potential effects in some of the experiments (Barnes 1972a).

Miller and others (1969) found that 3-year-old ponderosa pine seedlings sustained a 25-percent reduction in apparent photosynthesis after a 60-day exposure to an ozone concentration of 150 ppb for 9 hours per day. In another study, Coyne and Bingham (1981) measured changes in gross photosynthesis in needles of ponderosa pine trees of various sensitivities to ozone. Needles sustaining slight, moderate, and severe injury exhibited a 90 percent reduction in gross photosynthesis after exposure to a cumulative dose of 800,000, 700,000, and 450,000 ppb-hours ozone, respectively, in a 3-year time period (2 years for the most sensitive class of trees). The percentage inhibition in gross photosynthesis was based on photosynthesic rates of newly emerged needles; no true controls were used in the experiment. The authors emphasized that the decline in photosynthesis reflected the superimposition of ozone effects on normal aging.

Reich and Amundson (1985) over a period of several years measured the photosynthetic response of four tree species [sugar maple, eastern white pine, hybrid poplar (Populus deltoides x trichocarpa) and northern red oak] to ozone concentrations representative of those found in the ambient air of the Eastern United States. Exposure concentrations ranged from 20 to 140 ppb. A linear reduction in net photosynthesis was observed after long-term exposures (5 to 7 hour per day, 3 to 7 days per week, for 3 to 12 weeks) in all species. Reduction ranged from 10 percent in white pine at 30,000 ppb-hours to 40 percent in hybrid poplar at 20,000 ppb-hours. The authors suggest that, based on the results of their studies, reductions in net photosynthesis may be occurring over much of the eastern United States and southeastern Canada.

Botkin and others (1972) observed suppression of photosynthesis in eastern white pine when 5-year-old saplings were fumigated in small chambers. Three categories of sensitivity to ozone were observed. In sensitive trees, exposures of 900 to 1,000 ppb of ozone for 10 hours reduced net photosynthesis to zero; in trees of intermediate sensitivity, the same exposures reduced photosynthesis by approximately 50 percent; and in resistant trees, similar exposures had no effect upon photosynthesis. Ozone-induced suppression of photosynthesis in the trees with intermediate sensitivity was reversible if an ozone-free period followed exposure. Photosynthetic suppression and recovery occurred prior to or without the expression of visible injury symptoms.

d) Mixtures of Gaseous Pollutants

The effects of exposure to O_3, SO_2, NO, or NO_2 individually on photosynthesis in plants has been discussed above. The effects of mixtures of these substances on photosynthesis, however, have been poorly studied.

Sulfur dioxide, nitrogen oxides, and ozone individually inhibit the process of photosynthesis. Interaction of these chemicals frequently elicits a greater plant response than any of them alone do. The rate of reduction of net photosynthesis during the first 2 days of exposure was greater than additive when sugar maple and white ash were fumigated simultaneously with 500 ppb of SO_2 and O_3. For sugar maple, the reductions persisted for 1 week (Carlson 1979). The concentrations (500 ppb) used in this experiment are much greater than those usually encountered in the ambient air.

Fumigation of plants with high concentrations (1200 ppb) of NO_2 stimulates the formation of nitrate reductase (NiR). Plants exposed to NO_2 and SO_2 simultaneously failed to show an increase in NiR levels (Mansfield and Freer-Smith 1981). Wellburn (1982, 1985) has proposed a mechanistic explanation for the NO_2/SO_2 interaction. The presence of SO_2 completely prevents the induction of increased activity by NO_2. This phenomenon was observed in several clones of Lolium as well as in other grasses fumigated with low levels (68 ppb) of the two pollutants, while fumigation with NO_2 alone increased the levels of NiR (Wellburn 1982).

In a study of the effect of SO_2 and NO_2 in combination on alfalfa (Medicago sativa L.), White and others (1974) observed inhibition of the apparent rate of photosynthesis. Exposure to 2,500 ppb SO_2 for 2 hours resulted in 2 to 3 percent inhibition. Exposure to 2,500 ppb of NO_2 produced no measurable inhibition, but exposure to both gases simultaneously at 2500 ppb for 2 hours caused a 9 to 15 percent inhibition, while exposure to 15 ppb reduced photosynthesis 7 percent. The degree of synergism decreased as the concentrations increased until exposure concentrations reached 5,000 ppb. At that point, no synergism was observed. Bull and Mansfield (1974) also reported inhibition of photosynthesis in pea (Pisum sativum L.) by combinations of SO_2 and NO_2.

In the sunflower (Helianthus annuus L. cv Russian Mammoth), combinations of NO_2, SO_2 and O_3 measurably reduced the rate of photosynthesis when compared with the effects produced by each pollutant alone (Furukawa and Totsuka 1979). Inhibition of photosynthesis caused by $NO_2/SO_2/O_3$ combinations resembled those caused by SO_2/O_2 (Furukawa and Totsuka 1979).

Summary.--Photosynthesis in forest trees is altered by exposure to airborne chemical pollutants. Sulfur dioxide, nitrogen oxides, and ozone, as well as mixtures of the three chemicals, can inhibit the process of photosynthesis and decrease the amount of photosynthate produced. Regardless of the mechanism, a substantial reduction in photosynthesis will ultimately affect the growth, vigor, and reproduction of trees if repair does not occur.

4.2.4 Is Respiration in Forest Trees Altered by Exposure to Airborne Chemical Pollutants?

Respiration is the process through which the energy from the carbohydrates produced by plants during photosynthesis is released for growth, maintenance, and reproduction. In addition to its use in the formation of plant tissue and maintenance, some photosynthate is allocated for storage of compounds such as organic and amino acids, sugars, starches, fats, and proteins. Carbohydrates are also used to make secondary compounds such as alkaloids, tannins, pigments, and growth regulators (Waring and Schlesinger 1985). These processes occur at several sites within plant cells, and are likely to be vulnerable to disruption by gaseous air pollutants. Respiratory pathways and organelles may be affected (Black 1984).

Increased respiration is a well-recognized response to gaseous pollutant exposure (McLaughlin and Shriner 1980). Exposure to SO_2, O_3, or NO_x can alter such major physiological processes as photosynthesis in trees (section 4.2.3) and affect the rate of respiration. Respiration is sensitive to a variety of chemicals (e.g., SO_2) that may inhibit various stages of the overall process (Kramer and Kozlowski 1979). However, the conversion of carbohydrates, fats, and proteins into new plant tissue to repair pollutant-induced injuries requires large amounts of energy to be supplied through respiration (Kramer and Kozlowski 1979). Increased respiration, therefore, is caused by cellular injury rather than a cause of cellular injury (Kozlowski and Constantinidou 1986a).

Trees, when compared with plants in which foliage and seed production are predominant, require a disproportionate amount of carbohydrate for respiration because they have such a large amount of conducting tissue associated with their sapwood (Waring and Schlesinger 1985). Injury, therefore, requires changes in the normal allocation patterns and places stress on the starch reserves in trees. The maximum level of starch reserves in twigs and stems may serve as an indicator of the degree of injury a tree can tolerate (Waring and Schlesinger 1985).

Summary.--Increased respiration is a well-recognized response to gaseous pollutants. Energy normally used for maintenance is required to repair any injury caused by pollutants.

4.2.5 Are Carbon Uptake and Allocation Altered by Exposure to Airborne Chemical Pollutants?

Carbohydrates produced in the process of photosynthesis are normally used for the growth and maintenance of trees. Deciduous trees grow new leaves each year; conifers produce new needles. Roots, too, require energy to produce new, fine roots and develop mycorrhizae. Patterns of carbon allocation directly influence growth rate. The strategy for allocation of carbon may change during the life of the plant, as well as with different environmental conditions (fig. 27; Winner and Atkinson 1986). Factors such as air pollutants that limit carbon fixation shift allocation to new leaves, while factors that limit the availability of nitrogen or water will shift allocation to the roots. The increase in the allocation of photosynthate to new leaves due to the reduced photosynthetic capacity of plants exposed to ozone and to SO_2 has been reported by Oshima and others (1979) and by Reinert and Gray (1981). Therefore, air pollutants that limit carbon gain and nutrient availability may also suppress the growth rate and total biomass production (Winner and Atkinson 1986).

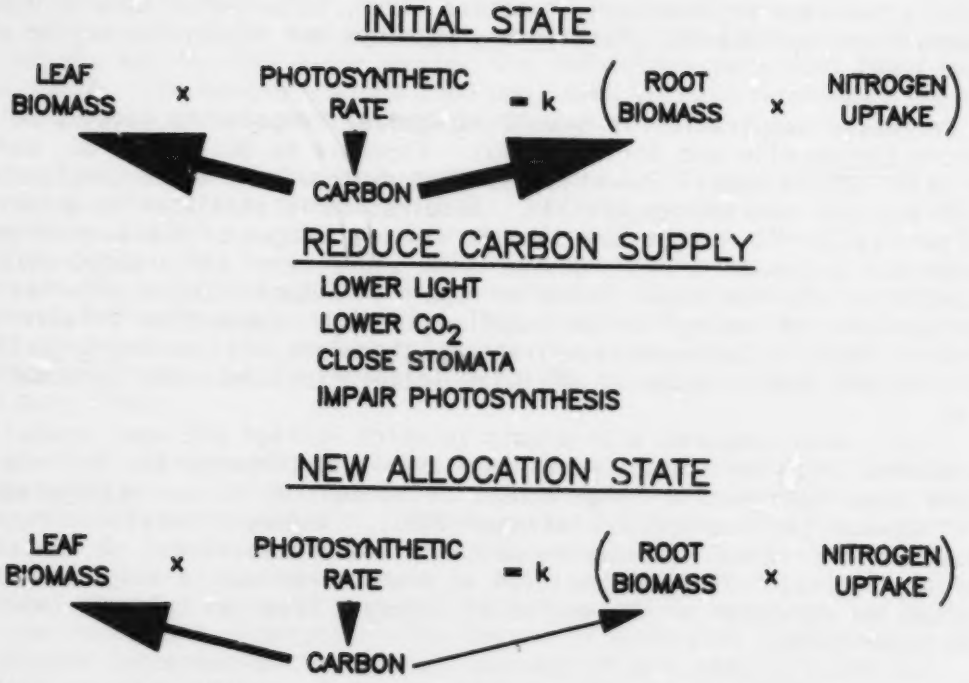

Figure 27. Model relating interaction between root and shoot allocation. Source: Winner and others (1985).

Changes in normal allocation patterns are an indication of stress (McLaughlin and Shriner 1980). Waring (1987) points out that the accumulation of amino acids in foliage is associated with reduced allocation of carbohydrates to the roots.

In the following section, changes in carbon allocation associated with experimental exposures to SO_2 and O_3 are discussed.

a) Sulfur Compounds.--Gaseous pollutants generally, and sulfur dioxide in particular, decrease the photosynthetic capacity of plants and shift carbon allocation towards the leaves. They also alter carbon translocation from the needles or leaves of trees. Suppression of root growth may result (Winner and Atkinson 1986). Inhibition of translocation of sugars by SO_2 without the accompanying inhibition of net photosynthesis also has been reported (Noyes 1980).

Inhibition of translocation was observed in bean (Phaseolus vulgaris L. cv. Black Valentine) when exposed to 100 ppb (26 $\mu g/m^3$) of SO_2 for 2 hours. Translocation was reduced 39 percent. Autoradiograms of ^{14}C-labeled primary leaves indicated that the phloem-loading processes and/or axial transport within sieve tubes were significantly inhibited or delayed by SO_2 exposure (Noyes 1980). Teh and Swanson (1982) also reported a reduction in phloem-loading. Translocation rates out of SO_2-stressed leaves were lower than expected on the basis of decreases in photosynthesis. Though photosynthesis recovers from SO_2 inhibition after exposures terminate, the effect of SO_2 on translocation persists (Darrall 1986; Noyes 1980; Teh and Swanson 1982).

Reinert and Gray (1981) observed changes in carbon allocation in radish (Raphanus sativus L.) when fumigated with 200 and 400 ppb SO_2 for 3 or 6 hours. A significant change in the root/shoot dry weight ratio was observed. Winner and others (1985) also reported a shift in carbon allocation from root to shoot in radish exposed to SO_2.

b) Nitrogen Compounds.--Reinert and Gray (1981) reported no change in root/shoot ratio in plants exposed to NO_2 at 200 and 400 ppb for either 3 or 6 hours. Though many studies cite the inhibiting effects of NO and NO_2 on photosynthesis, not many studies mention changes in carbon allocation resulting from the inhibition. However, the information regarding carbon allocation derived from studies using SO_2 and O_3 makes it possible to hypothesize that exposure to NO and NO_2 concentrations that inhibit photosynthesis does alter carbon allocation in trees.

c) Ozone.--Depression or inhibition of photosynthesis in the foliage can alter the allocation and translocation of photosynthate (e.g., sucrose) from the shoots to the roots and other organs (Tingey 1974). Tingey and others (1976a) observed that ozone exposure differentially affected the metabolite pools in the roots and tops of ponderosa pine seedlings grown in field chambers. The amounts of soluble sugars, starches, and phenols tended to increase in the tops and decrease in the plant roots exposed to 0.10 ppm O_3 for 6 hours per day for 20 weeks. The sugars and starches stored in the tree roots were significantly less than those in the roots of the controls.

McLaughlin and others (1982) observed that the decreased availability of carbohydrate in trees exposed to O_3 reduced the vigor of root systems and possibly enhanced the susceptibility of the trees to root diseases. The loss in vigor of the trees was accompanied by reduced annual radial growth and a loss in the capacity to respond in years when conditions were favorable for growth. The primary cause of decrease in radial growth appeared to be exposure to high concentrations of ozone and a sequence of events and conditions that led to premature senescence and loss of older needles, lower growth, and reduced photosynthate availability for growth and maintenance of trees (McLaughlin and others 1982). Carbon-14 transport patterns indicated that older needles were sources of photosynthate for new needle growth in spring and were storage sinks in the fall. The higher retention of ^{14}C-photosynthate by foliage and branches of sensitive trees indicated that the export of photosynthate to trunks and roots was reduced.

Data from herbaceous plants support the fact that O_3 inhibits the allocation and translocation of carbohydrates from shoots to the roots (U.S. Environmental Protection Agency 1986).

d) Mixtures of Gaseous Pollutants.--Reinert and Gray (1981) reported that O_3 exposure at 400 ppb reduced root dry weight more than exposure at 200 ppb during exposures of 3 or 6 hours. Sulfur dioxide also depressed the root weight at both concentrations (200 ppb and 400 ppb); however, when NO_2 and SO_2 were present together, there was a synergistic depression of the root/shoot ratio. The average O_3-induced reduction of root weight of radish was additive when NO_2 and SO_2 were present. Though the foliage was the direct recipient of the stress, the response appeared as a reduction in root weight. Exposures to pollutant mixtures do alter carbon allocation in herbaceous plants. If pollutant combinations inhibit photosynthesis and the production of carbohydrates, or alter biochemical or physiological processes in trees, changes in carbon allocation should result. The responses of trees to pollutant combinations and the effect on carbon allocation have not been studied.

Summary. Gaseous airborne pollutants, by decreasing the photosynthetic capacity of plants, shift the allocation of carbon into new leaves and away from the roots and also decrease the carbohydrates available for plant maintenance and storage. The amount of starch stored in branches and twigs may be an indicator of how much injury a tree can endure.

4.2.6 Is Increased Foliar Leaching in Forest Trees a Response to Airborne Chemical Pollutant Exposure?

Leaching of nutrients from the leaves of trees has been implicated as a result of ozone exposure. Leaching, a widespread natural phenomenon, is a biologically passive process that can remove a variety of substances from plants and temporarily reduce their concentrations in the tissues (Tukey 1970). Metabolites are leached by the action of aqueous solutions such as rain, dew, fog, or mist from the wide variety of plants that have been studied. Inorganic nutrients, organic substances, all of the naturally occurring amino acids, and many organic acids have been detected in leachates (Tukey 1980). The amounts of solutes leached can be greater than the amounts present in foliage prior to leaching, suggesting a connection with the transpirational stream. The rates of root uptake and translocation may increase during leaching (Tukey 1970; Tukey and others 1958).

Inorganic and organic ions are released from plant canopies during precipitation events and are a normal part of the forest nutrient cycle. Throughfall, the enhanced solute flux contained in water dripping directly from the canopy, and stemflow, the water that runs down the tree trunks to the forest floor, are normal features of the forest nutrient cycle and do not necessarily indicate injury from some extrinsic agent (Parker 1987). The materials released from plant canopies during precipitation come from two broad and not easily distinguishable classes of sources: 1) recycled materials, those originating from within the forest ecosystem; and 2) material chiefly of atmospheric origin from outside the system. Currently scientists are unable to distinguish the plant-derived contributions from atmospheric contributions to net throughfall (Parker 1987).

Enhancement of foliar leaching from forest canopies is most likely to be caused by deposition of materials from outside the system. Gaseous airborne chemicals could increase leaching by causing membrane or tissue injury. Little is known of the potential of gaseous airborne pollutants for increasing foliar leaching from forest canopies. These pollutants could enhance leaching by increasing the permeability of the plasma membrane, and by increasing the concentrations of certain soluble and potentially leachable metabolites such as carbohydrates and amino acids (Parker 1987). Currently neither SO_2 nor NO_x has been associated with increased leaching from forest canopies.

a) Ozone.--Ozone has been implicated as enhancing foliar leaching of nutrients by causing increased membrane permeability or alterations in cell metabolism. Krause and others (1984), in West Germany, associated limitations in growth of silver fir (Abies alba Mill.), Norway spruce [Picea abies (L.) Karst.], and certain hardwoods such as beech (Fagus sylvatica L.) with foliar injury. They fumigated seedlings with ozone concentrations ranging from 70 to 300 ppb in combination with acid fog, pH 3.5, continuously for 6 weeks. Krause and his co-workers reported that the entrance of ozone into the needles and leaves induced injury of the cell membrane and caused an uncontrolled loss of nitrate, Mg, Ca, K, and some sulfate. Leaching was enhanced by high light intensity and low nutrient supply in soils. Membrane injury occurred in the absence of visible injury. The authors suggested that the loss of nutrients and reductions in photosynthesis, carbohydrate production, and root growth resulting from ozone injury caused the trees to translocate nutrient reserves from older needles to the sites of greatest metabolic activity, the injured area. Altered carbohydrate allocation and nutrient uptake results in dieback.

b) Acidic Deposition.--Acids or acid-forming substances in wet and dry deposition could enhance leaching through ion exchange or erosion of wax and cuticle. These methods of enhancing leaching have been studied, chiefly in the laboratory or greenhouse.

The responses of forest trees to acid deposition are difficult to assess, though the processes of hydrogen ion exchange and weak base exchange provide mechanisms that could increase leaching, because the processes of throughfall enhancement make it difficult to attribute leaching to any one phenomenon. Processes capable of changing concentrations and amounts of precipitation are as follows (Parker 1987):

1) Atmospheric deposition may contribute basic materials such as soil dust, or acid-forming materials such as gaseous SO_2 and NO_x and sulfate aerosols;

2) The canopy may acidify precipitation through the release of weak, organic acids;

3) Canopy surfaces may increase pH by releasing basic cations in exchange for hydrogen ions in the incident precipitation.

Gaseous, particulate, and dissolved matter are potentially released from, taken up by, and transformed at, all canopy surfaces (Parker 1987). Because all of the processes cited above contribute to the alteration of precipitation quality, forest canopies commonly change the pH of precipitation reaching the forest floor. In addition, most of the field studies suffer because neither dry deposition inputs of hydrogen ions nor the stemflow releases of hydrogen and other cations are accounted for in the data. Moreover, highly acidic rainfall events, when compared with less acidic events at a given location, do not appear to enhance foliar leaching. Clearly, on the basis of the above reasons, change in pH alone is not a valid indicator that acidic deposition increases leaching.

A wide range of acidities (pH 6.7 to 2.3) has been used in laboratory studies of the effects of leaching. The degree of enhanced leaching is relatively small considering the range of acidities used. Moreover, the acidities used in the experiments are much greater than those normally encountered in bulk precipitation. No evidence exists for a pH level below which losses in solutes are dramatically increased (Parker 1987).

Summary. Foliar leaching is a widespread, natural, biologically passive phenomenon that is influenced by many external and internal factors. Gaseous, particulate, and dissolved matter are potentially released from, taken up by, and transformed at, all canopy surfaces. Release of materials from the forest canopy involves the action from two broad source categories: 1) from within the forest ecosystem, and 2) from without the ecosystem, chiefly from atmospheric deposition. At the present time ozone is the only gaseous airborne chemical that has been implicated as increasing foliar leaching, and then, when in association with acidic fog. The effects of acidic deposition are difficult to assess because forest canopies alter the pH of precipitation reaching the forest floor: 1) by contributing basic materials, or acid-forming materials (e.g., SO_2 and NO_x); 2) through the release of weak, organic acids; and finally, 3) by exchanging basic cations for hydrogen ions in incident precipitation. These processes of throughfall enhancement make it difficult to attribute leaching to a single phenomenon such as acidic precipitation. In addition, highly acidic rainfall incidents at a given location, when compared with those of low acidity, do not appear to increase foliar nutrient leaching.

4.2.7 Does Exposure to Airborne Chemical Pollutants Disrupt the Processes of Regeneration and Reproduction in Trees?

Reduction in the physiological efficiency of leaves often decreases crop yield and quality of seeds and fruits. Many studies indicate a reduction in yield in agronomic crops exposed to SO_2 and O_3 (see table 9 for critical

concentrations). Sensitivity to pollutants varies with the crop, cultivar, environmental conditions and with the concentration and length of exposure. Regeneration, reproduction, and yield are likely to suffer if pollutant exposures inhibit photosynthesis and the production of carbohydrates necessary for growth and maintenance. A few studies have shown pollutant inhibition of pollen germination and tube elongation under experimental conditions (fig. 28; Kozlowski and Constantinidou 1986a; Smith 1981).

Tree vigor is important in reproduction. In forest stands, dominant trees usually produce the most seeds. Dominant trees produced almost all of the cones in stands of ponderosa (Pinus ponderosa Dougl. ex Laws.) and sugar (P. lambertiana Dougl.) pine trees. Codominant trees produced only 1 to 1.5 percent, and almost no cones were produced by the intermediate and suppressed trees. In the crowns of red pine trees (P. resinosa Ait.), the most vigorously growing branches produced larger cones containing more and higher quality seeds than did the cones on the less vigorously growing branches (Kozlowski and Constantinidou 1986a). Finally, the importance of tree vigor to reproduction is illustrated by the fact that individual trees of loblolly pine (P. taeda L.) when released from competition (i.e., thinned) produced 10 times as many cones as did trees not released from competition (Kozlowski and Constantinidou 1986a).

Both regeneration and reproduction in trees can be affected by changes in carbon allocation (fig. 27). As indicated above, tree vigor is important for reproductive growth. Injury causes trees to shift their resources from growth, storage, and reproduction to injury repair. Defoliation is a classic example of a circumstance under which trees must regenerate tissue (McLaughlin and Shriner 1980). The ability of a tree to withstand such stresses can be assessed by evaluating the soluble and easily mobilized carbohydrate reserves near points of need. Webb (1981) reported that Douglas-fir [Pseudotsuga menziesii (Mirb.) Franco] and grand fir [Abies grandis (Dougl. ex D. Don) Lindl.] could be completely defoliated and still initiate new foliage, as long as starch concentrations in twigs remained above a certain minimum. The maximum amount of carbohydrate reserves in twigs and stems may be an indication of the degree of injury a tree can endure and still recover. Starch reserves are particularly important in deciduous trees that must survive much of the year on energy derived from storage (Waring and Schlesinger 1985).

a) Sulfur Compounds

Sulfur Dioxide.--Reduced production of cones in western larch (Larix occidentalis Nutt.), lodgepole pine (Pinus contorta Dougl. ex Loud), ponderosa pine (P. ponderosa Laws.) and Douglas-fir [Pseudotsuga menziesii (Mirb.) Franco] exposed to high concentrations of SO_2 was reported by Scheffer and Hedgcock (1955). Houston and Dochinger (1977) also reported significant differences in cone, seed, and pollen characteristics when 15-to 25-year-old eastern white pine and red pine (Pinus resinosa Ait.) growing in SO_2-polluted areas were compared with trees growing in SO_2-free areas. In white pine, percent pollen germination was reduced, seeds per cone were fewer in number and lighter in weight. In red pine, cone length, seed weight, percent filled seed, percent seed germination, and pollen tube length were significantly less in SO_2-exposed than in non-exposed trees. The reductions in cone, seed, and pollen production occurred despite the absence of visible injury to the trees.

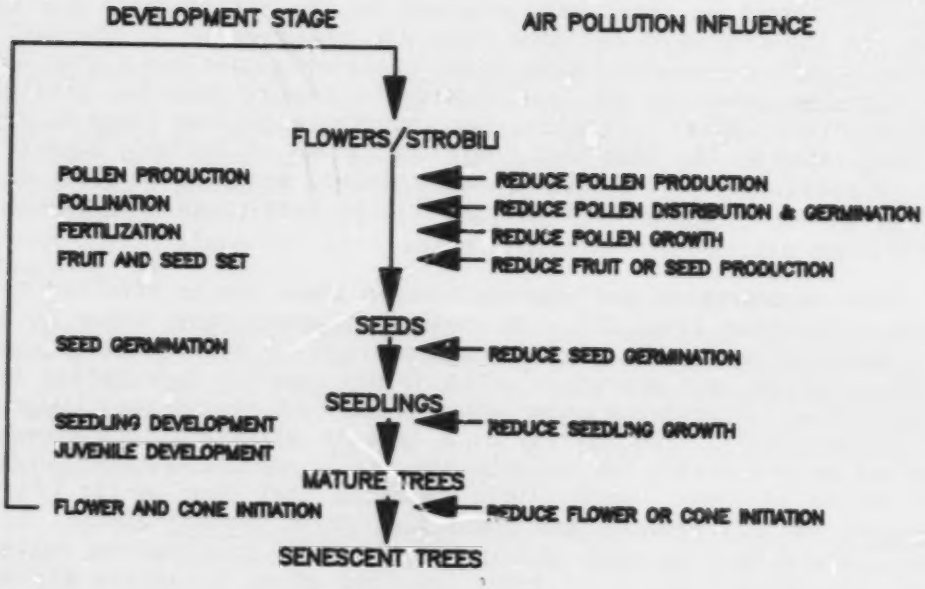

Figure 28. Potential points of interaction between air pollutants and sexual reproduction of forest trees.

Source: Smith (1981).

The effects on reproduction discussed above were observed on trees growing in the vicinity of power plants where SO_2 concentrations are usually high. A definitive statement concerning the effects of SO_2 at ambient concentrations on reproduction in forest trees cannot be made at this time.

b) Nitrogen Compounds

Nitrogen Oxides.--Nitrogen oxides have not been reported as altering reproduction and regeneration, probably because ambient concentrations do not usually reach injurious levels.

c) Ozone

Oxidant exposure of ponderosa and Jeffrey (Pinus jeffreyi Grev. & Balf.) pine in the San Bernardino forest of California was observed to reduce seed production. Dominant trees of both ponderosa and Jeffrey pines produced the most cones, and cone production increased significantly with tree age. However, dominant trees of both species 130 years or older, when severely injured by oxidant air pollution, bore significantly fewer cone crops over a 6-year period than did uninjured trees (Luck 1980). Miller and others (1982) suggest ozone injury-related reductions in photosynthesis, carbohydrate formation and allocation are the reason for reduced cone production.

In the eastern United States, reduction in cone or fruit production has not been reported even though ozone exposure has been reported to alter carbon uptake and allocation and to reduce vigor in sensitive trees (sec. 4.2.5). McLaughlin and others (1982) associated needle injury and senescence with a loss of vigor and reduced photosynthate availability in eastern white pine (Pinus strobus L.) trees exposed to ozone. Loss in vigor was accompanied by reduced annual radial growth and loss in capacity to respond to stress in years when conditions were favorable. Further, they state that repair of foliage injured by ozone exposure can place a drain on carbohydrate reserves. Moreover, a reduction in photosynthesis reduces the production of carbohydrate. Though the authors did not mention reduction in cone production, reduction in vigor and amount of photosynthate, as mentioned above by Luck (1980), affect reproduction.

Summary. Exposure to air pollution, specifically SO_2 and O_3, by inhibiting photosynthesis and altering the allocation of carbohydrates can decrease reproduction and influence regeneration in trees. Evidence from the ambient ozone exposure of ponderosa and Jeffrey pine in the San Bernardino Forest of California, as well as from experimental studies, indicates that dominant trees and trees with the most vigor produce the most cones. Trees in stands that were thinned produced more cones than trees not released from competition. The effects of gaseous pollutant mixtures on reproduction and regeneration have not been reported; however, if photosynthesis and carbohydrate production are reduced by exposure to gaseous pollutant mixtures, then reproduction is likely to be affected.

4.2.8 Is Host Susceptibility to Pests, Pathogens, and Mycorrhizal Colonization Altered by Exposure to Airborne Chemical Pollutants?

Plant tissue is generally not very nutritious to most insects and pathogens. Drought, air pollution and other environmental stresses, however, can

alter host susceptibility to attack from insect pests and pathogens by altering biochemical and physiological processes that influence the formation and concentrations of carbohydrates, nitrogen, sugars, and plant allelochemicals. Drought stress-related changes in nitrogen and carbohydrate-containing defense compounds can enhance the ability of insects to attack trees (Mattson and Haack 1987; Sharpe and others 1986; Waring and Schlesinger 1985). Similar changes have been observed in trees injured by air pollution. Mattson and Haack (1987) suggest that plants have a more or less universal response to stress. Insects appear capable of detecting and capitalizing on the changes caused by stress.

Tree vigor plays an important role in insect and pathogen attack (Hanover, 1975; Kozlowski and Constantinidou 1986b; Larsson and others 1983). A common observation is that vigorous standing trees of many different species are more resistant to bark beetle attack than freshly cut or lightning-struck trees (Hanover 1975). Larsson and others (1983) observed that ponderosa pine trees of low vigor were more prone to attack by beetles than trees of high vigor. Vigor was measured as stem growth per unit of leaf area. The attacks by beetles increased below a vigor threshold equivalent to production of approximately 100 grams of wood per square meter of leaf area. Susceptibility of lodgepole pine (Pinus contorta Dougl. ex Loud.) to mountain pine beetle (Dendroctonus ponderosa) attack also was found to increase below the same vigor index (Waring and Pitman 1985).

Christiansen and others (1987) hypothesize that the ability of trees to withstand attacks by bark beetles and their associated fungi is linked to the amount of carbohydrates that can be utilized directly for defense wound reactions. Therefore, any environmental factor that restricts the size of the canopy or its photosynthetic efficiency can weaken a tree's resistance.

To coexist, plants and parasites must be in a state of equilibrium in which neither totally dominates the other. Plants have evolved a variety of defense mechanisms, and parasites and pathogens have in return evolved countermeasures for avoiding or overcoming plant defenses. Neither plant defenses nor parasite countermeasures are completely effective; hence, a balance exists in which neither dominates. Air pollution-induced biochemical changes within plants can upset the relationship that the host and parasites or pathogens have developed over time (Hughes and Laurence 1984).

Different pine species may have evolved different defense strategies. Pines in the Southern United States have developed the resin ducts (constitutive defense) as contrasted with western conifers that have developed a hypersensitive wound reaction (induced defense). In the former, the defense system is present when the beetle attacks occur; in the latter, the defense system, a wound reaction, develops at the time of the attack. It has been hypothesized that the constitutive defense system evolved because southern trees experience asynchronous attacks by several multivoltine (having multiple broods in a season) bark beetle species. The resin duct system is the most energy efficient for them (Christiansen and others 1987). Air pollutants weaken and predispose trees to certain diseases, particularly those caused by non-obligate pathogens. Large infestations of bark insects were observed in forest stands that were weakened by air pollution (Hughes and Laurence 1984; Scheffer and Hedgcock 1955).

Plant parasites, specifically herbivorous insects, depend upon their host for nutrition and for many chemical and physical cues that regulate their feeding and reproductive behavior. Imbalances in the insect-tree relationship may be caused by variation in any of the four basic host characteristics: 1) morphology and anatomy of the host; 2) chemical repellents produced by the host; 3) cnemical attractants produced by the host; 4) nutritional status of the host (Hanover 1975). Biochemical changes induced by air pollutants can alter these relationships. The ensuing section cites some instances when this has occurred. Again, probably because their concentrations in ambient air are not high enough to cause injury to trees, studies mentioning the effects of nitrogen oxides in altering pathogen or parasite relationships do not exist.

a) Sulfur Compounds

Sulfur Dioxide.--Increased production of both chemical insect attractants and repellents has been reported in trees exposed to SO_2. Renwick and Potter (1981) reported increased emissions of the volatile terpenes that serve as chemical attractants for many insect pests after balsam fir [Abies balsamea (L.) Mill.] trees were exposed to SO_2 (200 ppb, 5 hours per day for 3 days). On the other hand, ethane, a strong repellent of the wood-boring beetle Monochamus alternatus, appeared after red pine (P. resinosa Ait.) and paper birch (Betula papyrifera Marsh.) were exposed to SO_2 at 300 ppb continuously for 3 to 5 days (Kimmerer and Kozlowski 1982). Production of oleoresin, another chemical repellent produced by trees injured by ozone, has not been reported for trees injured by SO_2. Sulfur dioxide exposure, however, increases the amounts of soluble sugars and nitrogen in plant foliage, inner bark, and sapwood of exposed trees (Mattson and Haack 1987).

Scheffer and Hedgcock (1955) observed increased numbers of bark beetles (Dendroctonus spp.) and engraver beetles (Ips spp.) on many of the larger ponderosa pines injured by SO_2 exposure. The authors suggest the attacks followed weakening of the trees and were not the primary cause of the weakening. Most species of bark beetles can only breed in trees that exhibit severe decline, or are already dead; therefore, they are merely promoting decomposition and mineralization. However, a few species are capable of attacking and killing living, sometimes quite healthy, trees (Christiansen and others 1987).

Air pollutants may increase or decrease the severity and incidence of plant disease depending upon whether it is the host or the pathogen that is adversely affected by pollutant exposure (Treshow 1980b).

Exposure to SO_2, according to reports, can suppress fungal growth. Scheffer and Hedgcock (1955) reported that fungi parasitic on needles and leaves appeared to be retarded or inhibited in areas exposed to the greatest concentrations of SO_2. Also, two species of rust fungi, Melampsora albertensis on quaking aspen (Populus tremuloides Michx.) and M. occidentalis on black cottonwood (Populus trichocarpa Torr. & Gray) were absent in areas with the highest SO_2 concentrations, sparsely present in areas where trees were moderately injured, and abundant where trees were uninjured. Linzon (1978) reported the virtual absence of white pine blister rust (Cronartium ribicula) from forests within 25 miles of the Sudbury, Ontario, Canada, smelters in line with the prevailing wind. A reduced incidence of fungi that attack plant foliage was also reported. In contrast, root-rotting and heart-rot fungi such

as Armillaria mellea, Glocophyllum abietinum (syn Lenzites abietina), Trametes serialis and T. heteromorpha were more common in trees injured by SO_2 than in uninjured trees.

b) Ozone

The majority of the studies relating the effects of ozone exposure on host-pathogen or insect-host relationship were made in the West where ozone injury of forests has been more prevalent.

Bark beetles are the most damaging and economically significant insect pests of commercially important conifers in the United States (Stark and Cobb 1969). Beetle outbreaks in western forests are associated with several predisposing factors. These include host weakening caused by photochemical oxidants; root disease initiated by the fungi Heterobasidion annosum (syn Fomes annosus) or Verticicladiella wagenerii (Stark and Cobb 1969); insect defoliation, such as pine looper stripping of ponderosa pine (Dewey and others 1974); and various climatic stresses, such as drought and windthrow (uprooting and breakage by strong winds) (Rudinsky 1962).

Ponderosa pines exhibiting advanced ozone injury symptoms were more frequently attacked by bark beetles than those with less severe symptoms (Stark and others 1968). The mountain beetle was not found infesting healthy trees. Ozone injury reduced oleoresin formation, exudation pressure, resin yield, and the rate of resin flow. The crystallization rate, however, increased. Further effects associated with ozone injury included reductions in sapwood and phloem moisture, thickness, and carbohydrates. Cobb and Stark (1970) concluded that the changes in host chemistry brought about by ozone injury made the trees more susceptible to bark beetle infestations. Mattson and Haack (1987) observed similar changes in oleoresin composition in trees under drought stress. They associated increased host susceptibility to insect attack with the changes.

Jones and Coleman (1988) and Coleman and Jones (1988a, 1988b) studied the interaction of plant stress and insect behavior and performance on eastern cottonwood (Populus deltoides Bartr. ex Marsh.) using ozone as the stress. Based on their studies, they conclude that an acute dose of ozone (200 ppb for 5 hours) that does not directly affect plant growth, or produce signs of visible injury, does not significantly change feeding and oviposition preference of the beetle Plagiodera versicolora. Both larvae and adult beetles preferred feeding on, and adults consumed more foliage of, ozone-stressed trees than controls when presented with a choice. In contrast, females preferred to oviposit on unstressed foliage. The changes in preference were maintained for 3 years and to approximately the same degree in two cottonwood clones.

The consequences of ozone stress on cottonwood and insect feeding and oviposition behavior are complex and not immediately obvious. To attempt a prediction of the effects of ozone stress and tree resistance to insect predation requires a comprehensive evaluation of the oviposition preference, studies on growth, survivorship, fecundity of the insects, their relationship to other members of the insect and pathogen community present on the cottonwood trees and, finally, the interaction of all of these organisms with the ozone-stessed trees (Jones and Coleman 1988). The authors suggest that the relationships of the cottonwood aphid (Chaitophorus populicola) and the leaf spot fungus

(Marssonina brunnea), which are unaffected by the same ozone exposures that affect the beetles and the cottonwood rust (Melampsora medusae) whose reproduction is reduced by those exposures, could be altered because of ozone stress having changed the leaf resource availability.

Mycorrhizae assist in protecting conifer roots from pathogens such as Heterobasidion annosum (Krupa and Fries 1971). Injury to the mycorrhizae or reduction in the number of mycorrhizae, such as can be induced by ozone exposure, can remove this protection. Non-mycorrhizal and mycorrhizal root systems contain essentially the same major volatile compounds; however, studies using Scots pine (Pinus sylvestris L.) indicate that the concentrations of monoterpenes and sesquiterpenes increase twofold to eightfold in roots infected by mycorrhizae. Many of the monoterpenes identified in mycorrhizal root systems are constituents of the oleoresins commonly found in conifers. Volatile oleoresin components from ponderosa pine have been shown to inhibit the growth of H. annosum and four Ceratocystis species (Cobb and others 1968), and are believed to aid in defense of tree from bark beetles (Stark and Cobb 1969). James and co-workers associated decreased oleoresin exudation with increased susceptibility to infection by H. annosum in roots (James and others 1980a) and cut stumps (James and others 1980b) of ponderosa and Jeffrey pines.

Christiansen and others (1987) also mention that production of resins, as well as phenolic compounds, in coniferous trees prevents fungal as well as beetle attack. When fungi, bacteria, or viruses infect plants, plants respond with a hypersensitive reaction. A necrotic area that deprives the invader of living tissues for food forms around the wound. If, due to the number of attacks, the resin concentration of the reaction zone decreases, the ability of the fungi to invade sapwood increases (Christiansen and others 1987). Skelly (1980) reported increased incidence of root disease caused by Verticicladiella procera in oxidant-injured eastern white pine in Virginia.

Air pollution injury and reduced tree vigor have been implicated as the predisposing factors in the infestation by the balsam wooly adelgid (BWA) of Fraser fir [Abies fraseri (Pursh.) Poir.] growing in the Appalachian Mountains. The Fraser fir in the Southern Appalachians is particularly susceptible to the BWA that arrived in this country from Europe around 1900. Atmospheric deposition is considered by Hain and Arthur (1985) to be the stress factor that alters the defense-response of Fraser fir and increases the host susceptibility to insect attack. Fraser fir decline on Mt. Mitchell, the highest peak in eastern North America, has in the past been attributed to the BWA. Recent observations suggest atmospheric deposition of SO_2, ozone, and/or airborne microscopic particles of metals (e.g., lead copper and nickel) probably play a role (Hain and Arthur 1985).

Tree mortality patterns of Fraser fir vary at the six major locations where they grow. Trees on Mt. Mitchell are more susceptible than trees growing farther north in Virginia on Mt. Rogers. Genetic differences and the greater ability of the trees on Mt. Rogers to produce monoterpenes have been associated with their greater tolerance to BWA attack. In experimental studies, monoterpene production between pre- and post-wounding control samples and the hypersensitive zone was more variable in Mt. Rogers trees. Terpene production in Mt. Mitchell trees, on the other hand, was stable. In addition, necrotic ectomycorrhizal roots were observed on the trees on Mt. Mitchell; this condition was not reported from trees growing on Mt. Rogers (Hain and Arthur 1985).

Mycorrhizae are sensitive to changes in the photosynthetic capacity of the host trees and their capacity to translocate carbon compounds to the roots (Hacskaylo 1973). For example, when seedlings of Virginia pine (Pinus virginiana Mill.), inoculated with the mycorrhizal fungus Thelephora terrestris and growing under a 16-hour photoperiod, were switched to 9-hour photoperiods, the seedlings became dormant within 4 weeks. No further invasion of rootlets by the fungus occurred even though root growth continued. Fungal sporophores were formed on the seedlings that remained under the 16-hour photoperiod. Studies have shown that simple sugars provided by plant roots are readily utilized by mycorrhizae and enhance fungal inoculation (Hacskaylo 1973; Krupa and Fries 1971). Berry (1961), in his studies to determine the cause of "emergence tipburn," dug up the roots of injured trees near Marlton, WV, where injury of eastern white pine trees had been prevalent for the past several years. The root systems of 10 healthy and 10 injured trees were examined. He observed that 72 percent of ozone-injured trees had dead feeder roots as compared with only 28 percent of the roots of healthy trees. Ozone exposure, therefore, not only inhibits the photosynthetic capacity of the eastern white pine, but also the altered carbon allocation is expressed as a change in the relationship between the roots and the mycorrhizal fungi.

Wargo (1972) reported that a reduction in the starch concentration in the roots of repeatedly defoliated sugar maple trees was related to a series of complex physiological changes that resulted in smaller amounts of sugars and changes in the number and concentrations of amino acids in the roots. These changes predisposed roots to attack by Armillaria mellea.

Ozone has also been shown to influence bacterial symbiosis in herbaceous species. Whether it does so in trees and woody shrubs appears not to have been investigated. In herbaceous species, the rate of nitrogen fixation by symbiotic bacteria is dependent on the rate of photosynthesis by the plant. Symbiotic nitrogen fixation is the major biological source of fixed nitrogen (Tingey and Blum 1973).

Ozone has inhibited infection of herbaceous plants by pathogens in controlled experiments; however, the inhibition occurred when ozone concentrations were several times greater than those usually encountered in the ambient air. Inhibition of forest pathogens by ozone has not been reported.

Summary.--Tree vigor is very important in host tree-insect interactions. Most species of bark beetles are able to breed only in trees that exhibit severe decline or are already dead. Air pollutants such as SO_2 and O_3, by altering the physiological processes within trees, tend to make them more susceptible to insect and pathogen attack. In addition, air pollutants can interfere with the natural defense processes within trees. Increased beetle and fungal attacks have been observed in forests subjected to SO_2 or O_3 exposure. The ability of trees to withstand environmental, insect, and pathogen attacks has been related to the amount of stored carbohydrate. It has been hypothesized that the ability of trees to withstand attacks by bark beetles and their associated fungi is linked to the amount of carbohydrates that can be utilized directly for defensive wound reactions. Therefore, any environmental factor that restricts the size of the canopy or its photosynthetic efficiency can weaken a tree's resistance. Air pollutants, by inhibiting the process of photosynthesis and the formation of photosynthate, can affect the tree's ability to withstand depredation by insects or pathogens.

A tree's ability to form mycorrhizal relationships has also been related to its vigor. Inhibiting photosynthesis and the amount of carbohydrate available for translocation to the roots can interfere with mycorrhizal formation, and thus further weaken the ability of trees to withstand environmental and biological stresses such as those posed by insects or pathogens.

4.3 Do Sulfur- and Nitrogen-Derived Pollutant Stresses Affect the Growth of Forest Trees?

Tree growth is the culmination of many biochemical and physiological processes. The carbon necessary for growth, maintenance, and reproduction of trees is obtained during the process of photosynthesis. Carbon dioxide diffuses from the atmosphere into the leaves of green plants through the stomata and into the intercellular spaces, then into the mesophyll cells where photosynthesis takes place. Within the chloroplasts in the mesophyll cells, the energy of sunlight is used to convert carbon dioxide into carbohydrates that then can be used in the other biochemical processes necessary for plant survival. Virtually every perturbation of a plant community results in stress and affects the performance and survival of individual plants. The discussions in the foregoing sections point out how exposure to airborne chemical stresses by affecting physiological and biochemical processes inhibit photosynthesis, alter carbon allocation, influence vigor and reproduction, and can increase susceptibility of trees to pathogens and insect pests. The impact that ambient air pollutants stresses (e.g., sulfur dioxide or ozone) have on growth is determined by the age of the tree, the complex interactions of pollutant and natural stresses with various environmental and genetic factors and their combined action on physiological and biochemical processes.

Because forest trees and shrubs are perennial plants, they must cope with the cumulative effects of both short- and long-term stresses. Responses of trees to some stresses may appear rapidly as, for example, when sensitive eastern white pine needles express visible symptoms within days after exposure to episodes of high ozone concentrations. In the majority of instances, however, responses are more subtle and may not be observable for many years. Adaptation and response of trees to stresses are expressed by differential growth, a result of changes in carbon allocation (Waring and Schlesinger 1985; McLaughlin and Shriner 1980). It is, therefore, usually difficult to attribute responses observed today to a stress or stresses that occurred many years previous. The sections that follow discuss studies in which air pollutants have been implicated as the cause of a reduction in tree growth.

a) Sulfur Compounds

Many observations under ambient field conditions and experimental studies indicate that exposure to SO_2 affects tree growth. The majority of the experimental studies used seedlings to study exposure-response because: 1) they require little space, are readily transported and grown, and can be easily exposed under controlled conditions; 2) they are highly susceptible; 3) they permit early detection of adverse effects; and 4) the experimental results are easily related to natural regeneration. Natural regeneration is important in forestry as an alternative to planting and in maintaining genetic diversity. A disadvantage, however, is that while the responses of older seedlings more closely resemble grown trees, the responses of young seedlings to air pollutants may not be applicable to large trees (Keller 1985). An

additional shortcoming of many of the studies is that the concentrations used usually were much greater than those encountered in ambient air. The advantage of using high concentrations, on the other hand, is that they permit the early detection of adverse effects, particularly visible effects.

In the studies that used concentrations approaching or equaling those encountered in the ambient air, foliar injury was usually the indicator of SO_2 exposure effects. Foliar injury, however, is not a reliable indicator that growth reductions have occurred or will occur. Growth reduction can occur without foliar injury (U.S. Environmental Protection Agency 1986). Carlson (1979) observed reduction in photosynthesis in sugar maple, black oak, and white ash without any foliar symptoms (table 12; sec. 4.3.2). Growth reductions were not mentioned, but the author suggests that plant productivity in polluted environments could be greatly reduced without visible symptoms appearing.

Only a few major investigations have reported observations based on trees growing under natural conditions (Dreisinger 1965; Dreisinger and McGovern 1970; Legge 1980; Legge and others; Linzon 1971, 1980; Scheffer and Hedgcock 1955; 1981). All of these studies were associated with point sources of SO_2 and were made in Canada. Scheffer and Hedgcock (1955) made the classical study associated with emissions from the smelters near Trail, British Columbia; Linzon and his co-workers reported on the Sudbury, Ontario area, while Legge and his co-worker studied the effects of SO_2 on a forest ecosystem near Whitecourt in west central Alberta.

In areas of moderate sulfur dioxide concentrations, Douglas fir and ponderosa pine saplings were reduced in vigor and many were killed. Near the smelters, the Douglas fir were most severely injured. The presence of SO_2 in the atmosphere resulted in the presence of abnormal concentrations of sulfur in the leaves (Scheffer and Hedgcock 1955).

Table 13 indicates the degree of injury to eastern white pine observed over a 10-year period (1953-1963) in the sulfur-fume-effects area of smelters near Sudbury, (Linzon 1980). Linzon (1971, 1978) indicated that a pollution (SO_2) gradient existed within the designated study areas. Effects correlated well with this gradient. Chronic effects on forest growth were prominent where SO_2 air concentrations annually averaged 17- or 45-ppb (45 or 115 $\mu g/m^3$) SO_2, and such effects were not reported in areas receiving only 8 ppb (21 $\mu g/m^3$) SO_2 annually. Although monitoring of SO_2 was conducted in these studies, neither concentrations of other air pollutants nor their potential interactions were evaluated by the authors. It is also not clear to what extent high, short-term, peak SO_2 exposures may have contributed to the effects that have been associated with annual average SO_2 concentrations.

In a study of the effects of low SO_2 concentrations on a forest ecosystem, the main ecological process directly and indirectly affected was nutrient cycling (Legge 1980; Legge and others 1980). The deposition of sulfur onto the soil progressively altered the mineral nutrient balances of ecosystem components by modifying the physiological and biochemical functions and relationships among the components. Laboratory and field measurements revealed a decrease in the rate of photosynthesis in lodgepole pine x jack pine (*Pinus contorta* Dougl. ex Loud. x *P. banksiana* Lamb.) hybrids in the

Table 13. The Degree of Injury of Eastern White Pine Observed at Various Distances From the Sudbury Smelters for 1953-63

Forest Sampling Station[a] (Distance and Direction from Sudbury)	Tree with Current Year's Foliage Injured in August 1963 (%)	Tree with 1-Year-Old (1962) Foliage Injured		Tree with 2-Year-Old Foliage		Net Annual Average Gain or Loss in Total Volume 1953-1963 (%)	Annual Average Mortality 1953-1963 (%)	Degree of SO_2 Damage	Average SO_2[b] Concentration for Total Measurement Period 1954-1963	
		June 1963 (%)	August 1963 (%)	Injured in June 1963 (%)	Lacking in August 1963 (%)				$\mu g/m^3$	(ppm)
West Bay (19 miles NE)	2.0	38.0	77.9	96.0	20.6	-1.3	2.6	Acute and chronic injury	115	(0.045)
Portage Bay (25 miles NE)	1.1	21.5	55.6	77.0	15.2	-0.5	2.5	Mostly chronic and little acute injury	45	(0.017)
Grassy to Emerald Lake (40 - 43 miles NE)	0.4	2.5	16.7	37.5	9.1	+1.8	1.4	Very little chronic injury	21	(0.008)
Lake Matinenda (93 miles W)	0.6	0.3	2.1	10.1	3.9	+2.1	0.5	Control: no SO_2 injury	3 (Sturgeon Falls)	(0.001)[c]
Correlation	0.96	0.96	0.93**	0.90**	0.94**	0.90**	0.81			

[a] Linzon (1971) (Pollutants other than SO_2 were not measured, and the monitoring was done several miles from the pine stands.)

[b] Dreisinger (1965)

[c] Data for 5-month growing season-1971

*$p < 0.05$

**$p < 0.10$

Derived from Linzon (1980).

field. Reduction of adenosine triphosphate (ATP) concentration in pine tissue during SO_2 fumigation in the field also was observed. Complete recovery followed termination of SO_2 fumigation. Basal area increment measurements of 200 lodgepole pine x jack pine trees from five ecologically comparable sampling sites revealed a decrease in wood production was directly related to the SO_2 concentrations. The maximum reduction in basal area increments occurred nearest the Whitecount Gas plant and fell to zero at 9.6 km. Recovery in the area began with the termination of SO_2 emissions. Again, these studies indicate greatest tree injury was observed near point sources and downwind from the SO_2 source (Legge 1980).

Reductions in tree growth observed after SO_2 exposures undoubtedly result from a reduction in the rate of photosynthesis and alterations in carbon allocation and translocation. These processes-together with other processes such as mobilization of storage reserves, reallocation of nutrients between old and new leaves, and the factors that regulate the distribution of plant nutrient resources between roots, shoots, and reproductive plant parts-influence the patterns of tree growth (Winner and others 1985).

b) Nitrogen Compounds

No published reports relating growth reduction of trees with exposures to NO or NO_2 in ambient air exist. Ambient air concentrations do not reach levels capable of causing injury to vegetation (National Research Council 1978b; U.S. Environmental Protection Agency 1982b). A study in California in the early 1970s compared the effects of NOx in smog with greenhouse fumigations (Thompson and others 1971). Leaf drop of navel orange trees was used to determine pollutant effect. Ambient concentrations of NO_2 in smog varied considerably, depending on the level of emissions, time of day, and the amount of sunlight. In early summer, peak concentrations of 120 ppb were observed; in late summer and early fall, peaks reached 240 ppb. The concentrations of other chemicals in the smog are not mentioned. Greenhouse fumigations were with 100 and 500 ppb. Exposures lasted 35 days. The authors concluded that "NO_2 levels in the Los Angeles Basin are not causing significant injury to citrus" (Thompson and others 1971).

Skelly and his co-workers (Skelly and others 1972; Stone and Skelly 1974) related reductions in growth of eastern white pine and yellow-poplar to emissions of NO_x alone or in combination with SO_2 from the U.S. Army Ammunition Plant at Radford, VA. These studies are the only published instances where injury to trees in the field have been associated with NO_x emissions. Sulfur dioxide, however, also was being emitted at the same time as NO_x. Since no air quality measurements were made during the period the growth reductions occurred, it is difficult to determine the concentrations of either pollutant at the time or determine which pollutant was causing the injury.

Munitions production levels at the arsenal were used as an indicator of the amounts of NO_2 and SO_2 being emitted because actual measurements were not available for the periods (during World War II, 1941 through 1945, and during the Korean conflict, 1950 through 1956) when the growth reductions occurred. Nor were they available for the period between World War II and the Korean conflict (1946-1949) when the tree growth increased. Tree growth was determined by using annual increment cores. Reductions in growth of eastern

white pine and yellow poplar were inversely correlated with production levels. High production rates were associated with decreased tree growth, low production levels with increased growth. Emission measurements were made during 1967 and 1969 at the time of the Vietnam conflict when production levels were high, as a possible basis for estimating the emissions during the years no measurements were made. Maximum 1-hour concentrations of NO_2 and SO_2 in 1967 were 120 and 40 ppb, respectively. For 1969, the maximum 1-hour NO_2 concentrations of 580 ppb and 670 ppb of SO_2 were substantially higher. Nitrogen dioxide concentrations for either year are far below those shown experimentally to cause vegetational injury (table 11; sec. 4.3).

Kress and others (1982) studied the growth impact of O_3, NO_2 and/or SO_2 on loblolly pine seedlings. Significant growth suppression was observed in the relatively sensitive full-sib loblolly pine families in all treatments except NO_2 alone. The concentration of NO_2 was 100 ppb. In another study, Kress and Skelly (1982) exposed willow oak, green ash, white ash, sugar maple, sweet gum, sycamore, yellow-poplar, pitch pine and loblolly pine seedlings to chronic doses of O_3 and NO_2. Height growth of loblolly and Virginia pine was suppressed by exposure to NO_2 at 110 ppb; however, white ash and green ash exhibited greater height growth than the control. In their final assessment, the authors state that "the relationship that data such as these have to ambient field conditions is tenuous."

The foregoing discussions of data reporting the effects of NO_2 exposure serve to reiterate the statement made earlier that concentrations of NO_x sufficient to injure trees growing in the forests are seldom if ever encountered except in the vicinity of point sources such as power plants or munitions factories.

c) Ozone

The effects of ozone on growth of vegetation, especially crop plants, are well documented and have been widely reviewed (Heck and others 1982; Krupa and Legge 1986; National Research Council 1977c; U.S. Environmental Protection Agency 1986). The effects of ozone on the growth of forest trees have not been studied as thoroughly. As in the case of SO_2, most of the studies cite the effects of ozone on seedlings. The majority of these experimental studies has exposed the seedlings in growth chambers or continuously stirred tank reactors; some have used open-top chambers in the field. Reich (1987) cites the results of many of these studies. The widespread distribution and regionality of ozone, however, have also made possible the observation of the effects of ozone exposure on trees growing in forests. The best documented studies are those in the San Bernardino Forest of California (Miller 1984; Miller and others 1982). There are, however, field studies and observations that indicate that ozone has been an important pollutant in the eastern United States for many years (Berry 1961; Berry 1964; Berry and Hepting 1964; Berry and Ripperton 1963; Hayes and Skelly 1977).

Generally, exposure to ozone is considered not beneficial to tree growth; however, growth stimulation of both herbaceous and woody plants by low ozone concentrations (20 to 50 ppb) has been reported (Bennett and others 1974; Kress and Skelly 1982). Growth stimulation of herbaceous plants occurred when plants grown in low ozone concentrations were compared with those grown in

carbon-filtered, ozone-free air. Bennett and others (1974) hypothesized that the apparent growth stimulation occurred because domestic herbaceous plants, such as the ones they used, were selected for low sensitivity to ambient ozone concentrations of 10 to 40 ppb. They were, therefore, not adapted to growing in an ozone-free atmosphere. Other studies also have observed that plants that are genetically adapted to growth in stressful environments frequently do not grow as well as when transferred to stress-free conditions (Roose and others 1982).

Kress and Skelly (1982) reported apparent growth stimulation when 2-to-4-week-old yellow-poplar (Liriodendron tulipifera L.), sweetgum (Liquidambar styraciflua L.), sycamore (Platanus occidentalis L.) and sugar maple seedlings grown in carbon-filtered, ozone-free air were compared with those exposed to 50, 100, and 150 ppb of ozone 6 hours per day for 28 consecutive days. Significant stimulation of growth in height of sweet gum, yellow poplar, and sycamore was observed at 50 ppb, and of sugar maple at 100 ppb. Growth suppression, however, was observed for sweetgum and sycamore at 100 ppb (see below). Kress and Skelly, when analyzing their results, stated that the relationship between their data and ambient field conditions was tenuous.

Growth suppression rather than stimulation is a much more common phenomenon resulting from ozone exposure. Under controlled conditions, Kress and Skelly (1982) observed significant suppression of height growth in 2- to 4-week old tree seedlings exposed to ozone for 6 hours per day for 28 days. At 50 ppb: loblolly pine (Pinus taeda L.) showed 18 percent reduction, American sycamore, 9 percent; at 100 ppb: pitch pine (P. rigida Mill.), 13 percent; sweetgum, 29 percent; green ash (Fraxinus pennsylvanica Marsh.), 24 percent; and at 150 ppb: willow oak (Quercus phellos L.) and sugar maple, 19 and 25 percent, respectively. Other investigators have reported similar results for other tree species exposed to ozone under other regimes (e.g., Constantinidou and Kozlowski 1979; Dochinger and Townsend 1979; Kress and others 1982; Mooi 1980; Patton 1981; Pye 1988).

Hogsett and others (1985), using exposures that simulated ambient conditions, noted a reduction in growth in height, diameter, and root length in two varieties of slash pine seedlings receiving chronic ozone exposure. The seedlings were exposed to one of three regimes: (1) charcoal-filtered air; (2) an exposure profile with a daily 1-hour maximum of 126 ppb at around 2 p.m., a 7-hour (9 a.m. to 4 p.m.) seasonal mean of 104 ppb, and an integrated exposure of 155,000 ppb-hour (sum of hourly ppm >0); or (3) a similar ozone exposure profile but with a daily 1-hour maximum of 94 ppb, a 7-hour seasonal mean of 76 ppb, and an integrated exposure of 122,000 ppb-hours. Both varieties of slash pine exhibited an increasing reduction in growth with increasing ozone concentration. A significant reduction ($p < 0.001$) in stem diameter occurred by day 112 for both ozone treatments: 24 percent less than that of controls for "elliottii" and 30 percent less for "densa" at the lowest O_3 exposure; and 40 percent and 50 percent below control plants for "elliottii" and "densa," respectively, at the highest O_3 exposure. Both O_3 exposures also caused significant reductions in growth in height ($p < 0.001$). The most pronounced change was observed in the growth of roots, which in "elliottii" was reduced 33 percent by day 21 at an integrated exposure of 29 ppm per hour, and 27 percent by day 56 with an exposure of 63 ppm per hour.

Studies dealing with the effects of ozone on growth of trees in their natural habitats have focused mainly on the San Bernardino forest ecosystem in California, where the effects of air pollution on forests were first observed. Studies of air pollution effects on forests in the Eastern United States have developed more slowly and, until reports (Johnson and others 1984; Johnson and Siccama 1984; Siccama and others 1982) of forest decline in the Northeast began to appear, were concentrated in the southern Appalachian Mountains, probably because air pollution injury of forests has not been a readily visible phenomenon.

Oxidant-induced injury of vegetation has been observed in the Appalachian Mountains for many years. The U.S. Forest Service in the 1950's studied the decline of eastern white pine in an area covering several hundred square miles on the Cumberland Plateau of East Tennessee, and concluded that unidentified atmospheric constituents were the cause (Berry and Hepting 1964).

Needle blight (also called emergence tipburn) of eastern white pine was first reported in the early 1900's, but it was not until 1963 that it was shown by Berry and Ripperton (1963) to be the result of acute and chronic oxidant exposure. An unusually high incidence of emergence tipburn of eastern white pine occurred during the summer of 1961 in western North Carolina and surrounding States. During the same period, researchers at North Carolina State University reported a very high incidence of weatherfleck of tobacco, a condition now known to result from ozone exposure (Berry 1961).

Investigations to determine the cause of the tipburn included examination of roots of injured and uninjured trees. On injured trees, 72 percent of the primary roots had dead ends; on healthy trees, 28 percent (Berry 1961). Associated with the injury were oxidant concentrations of 65 ppb for the period from 1:30 to 3:00 p.m. on July 1, and from 1:15 to 5:15 p.m. on July 2, 1961; earlier, on June 27, 50 ppb had also been measured in Pocahantas County, WV (Berry 1964). Measurements were made the following year to compare concentrations between the valley and mountain top. The highest concentrations were recorded during the day in the valley and at night on the mountain top. On the night of June 28, oxidant peaks of 220 ppb were recorded at 1:15 a.m., 120 ppb at 2:30 a.m., and 90 ppb at 8:45 a.m. For the valley, the highest concentrations (250 ppb) were observed between 12 noon and 5 p.m., during July 6 to 8, 1962. (Oxidant measurements were made with a Mast meter.)

Despite these early reports of Berry (1961, 1964), Berry and Hepting (1964), and Berry and Ripperton (1963), no concerted efforts were made to determine the effects of ozone on the vegetation of the Appalachian Mountains until Hayes and Skelly (1977) began monitoring total oxidants and reported oxidant-associated needle injury of eastern white pine at three rural Virginia sites between April 1975 and March 1976. The injury was associated with oxidant peaks of 80 ppb or higher. Hayes and Skelly suggested that continued exposure of sensitive and intermediately sensitive white pine to acute and chronic oxidant concentrations could ultimately influence their vegetative vigor and reproductive ability.

Needle injury symptoms and decreased growth of 25-year-old white pine growing on a plantation on the Cumberland Plateau near Oak Ridge, TN, were associated with ozone exposure (Mann and others 1980). To better understand

the mechanisms involved in growth reduction of the trees, Mann and others (1980) and McLaughlin and others (1982) conducted studies of the seasonal physiological responses of white pine. Several ozone episodes with 1-hour-average concentrations greater than 80 ppb were experienced between April 1 and July 17, 1977. A peak of 150 ppb was reached on June 16. Concentrations during the previous year (1976), however, had not been as high. Concentrations exceeded 80 ppb for only 15 days, as compared with 40 in 1977. Sulfur dioxide (100 ppb) and NO_x (200 ppb) levels during the study period were below those concentrations known to cause vegetational injury.

In a subsequent study of the same trees, McLaughlin and others (1982) found reduced annual radial growth, which they also attributed to high concentrations of ozone. McLaughlin and others (1982) divided trees into three sensitivity classes-tolerant, intermediate, and sensitive- on the basis of needle color and length, and duration of retention. A steady decline in annual ring increment of sensitive white pines was observed during the years 1962 through 1979. Reductions of 70 percent in average annual growth of sensitive trees, when compared to the growth of tolerant and intermediate trees, were noted. Tolerant trees showed a consistently higher growth rate of 5 to 15 percent ($p = 0.005$) than intermediate trees for the 1960 to 1968 interval, similar growth from 1969 through 1975, and reduced growth of 5 to 15 percent (but significant only at $p = 0.10$) for the period 1976 through 1979, compared to trees of intermediate sensitivity. Needles of ozone-sensitive trees were 15 to 45 percent shorter than those of either of the other classes. Decline was attributed primarily to chronic exposure to ozone, which frequently occurred at phytotoxic concentrations in the area. For the years 1975 through 1979, the incidence rates for hourly concentrations $\geq$80 ppb were: 1976, 190 hours; 1977, >339 hours; 1978, 190 hours; 1979, 125 hours. Maximum 1-hour concentrations ranged from 120 to 300 ppb during this period. The pollutants sulfur dioxide and fluoride have been measured in the area, but the premature loss of needles and occasional tip necrosis of needles of the current year are manifestations usually associated with ozone, and no cause-and-effect relationship with sulfur dioxide is indicated by the available data.

In the studies discussed above (Mann and others 1980; McLaughlin and others 1982), decline in vigor and reduction in the growth of coniferous trees were usually associated with the following sequence of events and conditions: (1) premature senescence and loss of older needles at the end of the growing season, probably leading to (2) reduced carbohydrate storage capacity in the fall and reduced resupply capacity in the spring to support new needle growth; and (3) increased reliance of new needles on self-support during growth. This resulted in (4) shorter new needles, lower gross photosynthetic productivity and (5) a higher retention of current photosynthate by foliage, thus reducing the availability of photosynthate for external use (including repair of chronically stressed tissues of older needles) (McLaughlin and others 1982).

Benoit and others (1982) studied the annual ring increment of native eastern white pines of reproducing age to evaluate the possible effects of oxidant air pollution on the long-term growth of forest species in a region of the Blue Ridge Mountains of Virginia, extending from the northern end of Skyline Drive in Shenandoah National Park to the southernmost portion of the

Blue Ridge Parkway lying in Virginia. The three white pines in each study plot were classified as sensitive, intermediate, or tolerant, based on a foliar rating scale that incorporated needle length, needle retention by number of years, and the presence of typical ozone symptoms on needles. The mean ages of ozone-tolerant, intermediate, and ozone-sensitive tree classes were 53, 52, and 56 years, respectively. From 1955 to 1978, growth in mean annual radial increment for sensitive trees was 25 percent less, and for intermediate trees 15 percent less than that of tolerant trees, respectively. Only the 25 percent decrease for ozone-sensitive trees, however, was significant ($p = 0.01$; table 14). Smaller mean increments in the last 10 years compared to the previous 24 years indicated a trend toward decline in overall growth rates in all classes of trees. A comparison of growth from 1974 to 1978 with that from 1955 to 1959 showed decreases of 26, 37, and 51 percent for tolerant, intermediate, and sensitive trees, respectively.

The significant reduction in radial growth of ozone-sensitive white pines was assumed by the authors (Benoit and others 1982) to be the result of cumulative stress caused by the reduced photosynthetic capacity of oxidant-injured trees. Their assumption was based on the fact that no significant changes in seasonal precipitation had occurred between the 1955-1963 and 1963-1978 periods. Increasing ozone concentrations, therefore, could account for growth reductions, particularly since for the latter period there was a negative correlation between precipitation and radial growth. Sulfur dioxide concentrations were too low (undetectable to 30 ppb) to have any vegetational effects.

Duchelle and others (1982) reported that during a typical ozone season concentrations increased from January through June, stabilized during the summer months, and then decreased around October. During this time, ozone concentrations are low at night, begin to rise in the morning, peak in early afternoon, and then drop again at sunset. Episodes in the Blue Ridge Mountains lasting from 1 to 5 consecutive days have been reported by Skelly and others (1984) (table 15). Monitoring of ozone indicated the presence of monthly average concentrations of 50 to 60 ppb on a recurring basis with episodic peaks (1-hour) frequently in excess of 90 ppb (Duchelle and others 1982).

Earlier, Hayes and Skelly (1977) suggested that the episodes in the area resulted from long-range transport of ozone from urban areas. This suggestion was supported by Fankhauser (1976), whose observations indicated that ozone was transported in a giant loop stretching from New York City, Philadelphia, Baltimore and Washington, DC, though West Virginia and Ohio and then back to the Wheeling, WV-Pittsburgh, PA, area. Ozone was observed to follow this path for 4 to 5 days in September 1972. Earlier, in May 1972, another instance of ozone transport had been observed when a stagnant "high" and a slow-moving "low" transported air from Chicago and Pittsburgh areas southward along the Appalachian Mountains to the Gulf of Mexico and then back to Miami, FL.

The experiments of Duchelle and others (1982, 1983) exposing native tree seedlings and native vegetation in the Big Meadows area of the Shenandoah National Park in the Blue Ridge Mountains of Virginia to ambient ozone, indicate that ambient concentrations can reduce both the growth of native trees other than eastern white pine and the productivity of the native herbaceous vegetation found growing in the forested areas.

Table 14. Annual Mean Radial Growth Increment, 1955 Through 1978,[a] for Three Ozone Sensitivity Classess of Native Eastern White Pines (Pinus strobus L.) Growing in 10 Plots of Three Trees Each in the Blue Ridge Moutains in Virginia

	(mm)		
Plot	Tolerant Trees[a]	Intermediate Trees	Sensitive Trees
1	4.59	2.13	3.08
2	3.52	2.12	2.86
3	8.19	6.34	6.89
4	4.80	3.75	2.62
5	5.94	6.53	5.73
6	4.64	3.76	2.62
7	2.85	2.75	1.51
8	3.91	4.52	1.96
9	3.32	2.04	2.61
10	1.67	2.98	1.46
Mean	4.34 A[b]	3.69 AB	3.10 B

[a]White pines rated tolerant, intermediate, or sensitive to O_3 based on foliar symptoms.

[b]Sensitivity classes with the same letter are not significantly different at p = 0.01 based on Duncan's multiple range test.

Source: Benoit and others (1982).

Suppressed growth of seven indigenous forest tree species was observed when seedlings grown in open plots or ambient air open-top chambers were compared with those grown in charcoal-filtered air (Duchelle and others 1982). Growth reductions of wild-type seedlings of tulip poplar, green ash, sweet gum, black locust (Robinia pseudoacacia L.), eastern hemlock [(Tsuga canadensis (L.) Carr.)], Table Mountain (Pinus pungens Lamb.), Virginia (P. virginiana Mill.) and pitch (P. rigida Mill.) pines usually occurred without expression of foliar symptoms. Open-top chambers were operated continuously in 1979 from May 9 until October 9, and in 1980 from April 24 until September 15.

Injury to and growth reductions of native herbaceous vegetation growing in the Big Meadows area also were observed (Duchelle and others 1983). Few studies have reported the effects of ambient air pollutants on growth and productivity of native vegetation. Reduced growth and productivity of graminoid and forb vegetation were observed in experiments conducted at the same time and under the same exposure conditions as the tree seedling study cited above. Biomass production (weight of living tissue) was greatest in filtered open-top chambers when compared to those in open-air plots. Total 3-year cumulative dry weight of plants in filtered chambers was significantly greater ($p = <0.05$) than that of plants in non-filtered and open-air plots. Reductions in biomass were attributed to a decrease in root growth, a result

Table 15. Peak Hourly Ozone Concentration in Responses Recorded at Three Monitoring Sites in Western Virginia, Spring and Summer, 1979 Through 1982

Date	1-hr O^3 concn., ppb		
	Big Meadows	Rocky Knob	Horton Center
1979, April 11-12	100	128	N.A.[a]
1979, June 5-6	082	112	N.A.[a]
1979, September 12	095	079	072
1980, May 29-30	093	088	063
1980, June 13-15	093	088	063
1980, July 14-15	089	080	N.A.[a]
1980, July 31	102	113	129
1981, May 24	084	054	094
1981, June 23-23	N.A.[a]	106	073
1981, June 29-30	N.A.[a]	090	072
1981, July 8-11	096	129	114
1982, May 11-13	125	N.A.[a]	095
1982, May 16-17	090	N.A.[a]	085
1982, July 24-26	110	080	125
1982, July 27-28	090	080	110
1982, August 3-4	090	075	115
1982, August 19-20	095	065	090
1982, October 1-3	100	075	080

[a]N.A. = data not available.

Source: Skelly and others (1984).

of ozone exposure. Common milkweed (Asclepias syriaca L.) and common blackberry (Rubus allegheniensis Porter) were the only two native species to develop visible injury. Milkweed previously had been reported to be sensitive to ozone and a good biological indicator (Duchelle and Skelly 1981).

In the introduction to this section it was pointed out that perennial trees and shrubs must cope with the cumulative effects of both short- and long-term stresses, and that responses to these stresses may be subtle and not observable for many years. McLaughlin and others (1982) associated long-term (1962-1979) growth reductions of eastern white pine with ambient ozone concentrations by using data from increment cores. Benoit and others (1982) likewise used increment cores to relate growth reduction of eastern white pine in the Blue Ridge Mountains during the years 1955 to 1978 to ambient ozone concentrations. More recently, dendroecological studies of dieback and decline of red spruce (Picea rubens Sarg.) in the Northeast (Johnson and Siccama 1983) and of reduced growth rates of red spruce, balsam fir (Abies balsamea (L.) Mill.), and Fraser fir [(A. fraseri (Pursh) Poir.)] in central West Virginia and western Virginia (Adams and others 1985) have provided further evidence that growth reductions measurable today probably began at least 20 years ago. Currently, there is no agreement as to the factor or factors that precipitated the dieback, mortality, and decline in growth, but multiple stresses including air pollution have been hypothesized (Adams and others 1985; Johnson and Siccama 1983).

The dendroecological studies of Phipps and Whiton (1988) also evidence the difficulty of determining the cause today of tree growth responses that began many years ago because of a major stress or stresses existing at that time. Growth decline, defined as actual growth less than that expected, was observed in 40 of 60 tree-ring collections judged to have been unaffected by local site histories, from 89 locations throughout much of the range (from Connecticut to North Carolina and west to Iowa) of white oak (Quercus alba L.). Onset of decline began somewhere between the mid-1950's and the early 1960's, and was essentially the same in the Northeast, Midwest, and the Southeast. The chronologies studied were of trees that had reached canopy dominance by 1850; some individuals were more than 200 years old at that time. A linear growth trend prior to 1950, as determined by examination of the basal area index (BAI), was observed in these dominant trees. A number of the individuals were cross-dated as having been in the canopy before 1600. Studies of the BAI of these individuals indicate no departure from the observed linear trend prior to 1950.

The onset of growth decline, though rather abrupt, did not appear in all trees at the same time, suggesting that some trees were more tolerant of the stress than others. Some trees showed no decline at all. Changes in climate did not appear to mimic growth trends except in North Carolina where a temperature maximum was observed around 1950. Estimations of long-term regional sulfur and nitrogen trends vary for the different regions (Phipps and Whiton 1988). Trends in the Southeast generally show a marked increase in sulfur emissions since 1950, and those in the Midwest appear to have been increasing since the 1920's. In the Northeast, trends in sulfur emissions have stabilized or decreased somewhat since the 1970's while nitrogen emissions have increased. Only in the Southeast have emission increases correlated with the decline in white oak growth; however, trees that did not exhibit a decrease in growth in the 1950's still do not exhibit a decrease even though emissions have increased. The authors hypothesize that growth decline was initiated during a period beginning in the mid-1950's to the early 1960's by conditions or events as yet unidentified, and that subsequent growth has not been in response to tree age, geographic location, site quality, climate trends, or regional emissions of sulfur and nitrogen oxides (Phipps and Whiton 1988).

Phipps and Whiton (1988) rule out sulfur and nitrogen emissions as initiators of the growth decline observed in white oak since the 1950s. Could ozone have been an initiator of the decline? The authors do not speculate. It should be noted, however, that regular oxidant-monitoring stations were not established east of the Mississippi River until 1962 (U.S. Department of Health, Education, and Welfare 1970). Maximum oxidant concentrations recorded between 1964 and 1967 indicate that Cincinnati had 10 days above 150 ppb and 55 above 100 ppb; Philadelphia 13 days above 150 ppb and 60 above 100 ppb; and Washington, DC, 7 days above 150 ppb and 65 above 100 ppb. Based on these data, ozone concentrations sufficient to injure vegetation were already present in the atmosphere from Chicago to Philadelphia and Washington, DC, by the middle 1960's. The observations of Berry (1961) and Berry and Ripperton (1963) mentioning oxidant concentrations above 100 ppb during 1961 and 1962 in West Virginia and North Carolina indicate that the area with high ozone concentrations also extended southward. Oxidant exposure at this time was just being established as a probable cause of the foliar injury that had been observed for many years on sensitive eastern white pine (Berry and Ripperton 1963).

d) Mixtures of Gaseous Pollutants --

The possibility that exposure to mixtures of gases could have an effect on vegetation greater than a single gas has been a concern since the 1960's. A majority of the studies conducted since that time has focused on the effects of pollutant mixtures on agricultural crops, and the SO_2/O_3 combination has received the greatest attention.

Dochinger and others (1970) suggested that O_3/SO_2 acting synergistically could cause needle mottling of white pine. Karnosky (1976) observed more than additive effects when he exposed trembling aspen (Populus tremuloides Michx.) to combinations of O_3/SO_2. He also observed clonal variation in the response of the different clones to the gas combinations. Clones shown to be sensitive to SO_2 or O_3 alone were more sensitive to the combinations of the two. These assessments were based on leaf injury rather than growth response. Both of the above studies were conducted in chambers.

Miller and Stolte (1984) exposed Jeffrey (Pinus jeffreyi Grev. & Balf.), ponderosa (P. ponderosa Dougl.ex Laws), and Digger pine (P. sabiniana Dougl.) to mixtures of SO_2/O_3. Shoot growth was not as sensitive as root growth. Root growth was more affected by O_3/SO_2 in combination than by either pollutant alone.

The co-occurrence of SO_2 and NO_2 is most frequently measured in the vicinity of power plants (Lefohn and Tingey 1984). Ambient concentrations of NO_2 rarely approach the injury threshold. Concern regarding its presence in the atmosphere is related to its potential for interaction with other pollutants such as SO_2 as demonstrated by Tingey and others (1971). The majority of the experiments conducted to determine the effects of $SO_2 + NO_2$ in combination have used agricultural and horticultural crops.

In England, Whitmore and Freer-Smith (1982) and Freer-Smith (1984) observed that combinations of $SO_2 + NO_2$ affected cuttings of a number of British tree species. Second year cuttings were exposed to SO_2 and/or NO_2 from March through August. The $SO_2 + NO_2$ combination (62 + 62 ppb) produced interactive effects resulting in greater than additive responses. Significant reductions in stem height and diameter and dry weight resulted from the exposures (Whitmore and Freer-Smith 1982). Freer-Smith (1984) also reported that decreases in growth produced by $SO_2 + NO_2$ were greater than by either pollutant alone.

Stone and Skelly (1974) and Phillips and others (1977a,b) measured growth responses in the field to emissions from the Radford Army Ammunition Plant in Radford, VA. Both SO_2 and NO_2 were present in concentrations sufficient to influence the growth of white pine, yellow poplar, and loblolly pines. No attempt was made at this time to see whether the pollutant combination potentiated the effect of either pollutant alone; however, synergism was suggested.

The O_3 and NO_2 combination has been studied infrequently. Kress and Skelly (1982) exposed seedlings of 10 forest tree species to O_3 and NO_2 concentrations of 100 ppb. The only significant interaction effects noted between the two pollutants were less than additive.

The growth impact of O_3, NO_2, and/or SO_2 on loblolly pine was studied by Kress and others (1982). Adding NO_2 to the O_3 and SO_2 combination had a slightly stimulatory effect on growth, however, the "sensitive" seedling of the full-sib family suffered a 30 percent height suppression when exposed to the three-pollutant combination as compared with a 14-percent suppression for the resistant family.

Summary. Evidence that exposure to sulfur oxides and ozone can decrease the growth of trees is well established by the many experimental studies that have been conducted during the past 20 years. Evidence also is strong that the growth of trees in their natural habitats also can be inhibited by exposure to these two pollutants. Sulfur dioxide inhibits growth and causes decline and mortality of trees in the vicinity of point sources. Ozone also inhibits growth and causes decline and mortality in areas where episodic concentrations (frequently transported from metropolitan areas some miles away) last for several days and reach 80 ppb or higher. Data indicating nitrogen oxide injury of trees growing in their natural habitats are lacking. The concentrations used to cause injury experimentally have not been observed in the field.

One of the biggest problems facing scientists at the present time is determining what stress or stressors precipitated tree injury or growth responses visible or measurable today when the exposure took place a number of years ago. Also, though sulfur dioxide and ozone do inhibit tree growth, the exact manner in which they alter tree biochemistry is as yet unknown, and as yet it is not possible to establish an exposure-response relationship.

4.4 What is the Likelihood That Airborne Chemical Pollutants Affect Forest Ecosystems Through Soil-Mediated Response Mechanisms?

Soils are one of the most stable components of ecosystems, and together with climate determine the limits of ecosystem productivity. Soils are a mixture of inorganic minerals, organic matter, soil animals, microorganisms, gases, and water. They function as the anchor for most terrestrial and many aquatic plants. They provide and store water, nutrients, and oxygen needed by plant roots for growth. Chemical transformations and animal and microorganismal activity within soils influence the availability of nutrients to plants. Should airborne chemical pollutants inhibit or alter chemical processes, or influence animal and/or microorganismal behavior in soils, the growth of the above-ground flora could be impacted.

Exposure of plants to airborne gases, fine aerosols, or coarse particles through the soil is indirect. To have an effect, these chemical compounds must in some way influence the movement or uptake of plant nutrients. Four separate processes are involved in exposure: 1) deposition, 2) dissolution into soil water, 3) percolation or diffusion through the soil, and 4) absorption from the soil through the surfaces of fine feeder roots and mycorrhizae. Many biotic, physical and chemical factors influence these processes.

The general consensus of most investigators at the present time is that there are no apparent drastic or potentially catastrophic short-term effects from the atmospheric deposition of ambient gaseous air pollutants on soils. There is a lack of information, however, concerning either temporary or

permanent effects of long-term pollutant exposures on soils (Weidensaul and McClenahen 1986). Most of the available information concerning pollutant-soil interactions deals with soils as a sink. For this reason, the effects of deposition and fate of gaseous airborne chemical pollutants on soils are discussed in a very general manner.

The effects of acidic deposition on soil are of greatest concern. A number of publications have reviewed this topic (Altshuller and Linthurst 1984; Bennett and others 1985; Binkley and others 1987; Brandt 1987). These publications discuss in some detail the possible impact on soils of acidic deposition and the resulting effects on vegetation changes. The following section deals with the relative likelihood of some of the major hypothetical mechanisms by which soil changes could affect forest plants.

4.4.1 Airborne Gases, Fine Aerosols, and Coarse Particles

Soils generally sorb organic gases faster and in greater amounts as their molecular weights increase. Absorption rates also increase when functional groups such as nitrogen, oxygen, and sulfur are attached to the organic gases. Uptake of organic gases usually depends on the development of an appropriate microorganismal population, while removal of inorganic gases is attributed to chemical and/or physical processes (Bohn 1972).

Most gaseous SO_2 is removed by soil within 15 minutes, but the same amount of NO_2 requires approximately 24 hours. The sorption rate for each of the compounds is similar in sterile and nonsterile soils, suggesting that most SO_2 and NO_2 is removed by some chemical reaction (Abeles and others 1971). The magnitude of SO_2 and NO_2 sorption is influenced by soil type. Soils not only sorb NO_x gases, but also release nitrogen gases such as N_2, N_2O, O, NO, and some NO_2 via chemical reactions (Bohn 1972).

Nitrates and sulfates diffuse rapidly into the soil. Length of nitrate retention depends on their assimilation by plants and microorganisms, as they are readily leached. The ammonium ion (NH_4^+) is frequently attached to nitrates or sulfates. Nitrates, sulfates, and ammonium when in ionic form in the soil are readily taken up by tree roots (Donahue and others 1977). Retention of $SO_4^=$ depends on soil pH and the presence of hydrous oxides, iron, and aluminum (Abrahamsen and Dollard 1979). Acidic nitrates and sulfates as aerosols or in rain tend to acidify soil. The nutrient roles of sulfur and nitrogen are mentioned in section 4.3.1; the effects resulting from deposition in acid aerosols are discussed below.

Soils have developed a nutrient capacity during their formation from parent materials over time. Trees and other vegetation growing on these soils are adapted to them. Forests, therefore, are not nutrient limited, except for a lumberman's point of view. Today, as a result of atmospheric pollution many coniferous forest soils are receiving nitrogen in excess of the growth requirements of the trees. Appearance of nitrate in the soil solution is an early symptom of excess nitrogen in these systems. A second symptom, because of the inability of roots of the densely growing conifers to metabolize large amounts of nitrate, is the presence of nitrate in the xylem sap (Waring 1987).

Nitrogen uptake by trees may not necessarily enhance growth, particularly in boreal and subalpine conifers. The photosynthetic capacity of conifer

foliage is low and not greatly enhanced by increasing the nitrogen content. In addition, excess nitrogen not only alters the way in which carbon is allocated between shoots and roots, but also is in a form difficult to metabolize, and increases sensitivity to frost. The capacity of gymnosperms in general, and boreal and subalpine species in particular, to synthesize the enzymes required to reduce nitrate in foliage or roots appears to be limited. A reduced allocation of carbohydrates to the roots is associated with the accumulation of amino acids in foliage (Waring 1987).

Excess nitrogen in subalpine or boreal ecosystems is likely to cause nutrient imbalances and make conifers more susceptible to drought, other pollutants, and pathogens. Competition under these circumstances would favor deciduous species (Waring 1987). Whether nitrate deposition in the Southeastern United States is capable of causing the phenomena presented above has not been assessed at this time.

4.4.2 Acidic Deposition

Acidification can strongly influence soil dynamics by altering the soil's biological, chemical, and physical behavior. Changes in soil structure, soil aeration, water status, and the environment of soil organisms can, in turn, influence the availability of nutrients and toxic substances. Soil acidification can result from natural sources of hydrogen ion in soils as well as from the atmospheric deposition of acidifying substances (Brandt 1987). Sulfate and nitrate deposition are expected to have a greater influence on most soils than hydrogen ion deposition (Bennett and others 1985). If the sulfate and nitrate in acidic deposition are not assimilated by plants and microorganisms, the addition of the hydrogen ion could increase soil acidity and lower soil pH (Binkley and others 1989).

Effects of soil acidification in general may be outlined as follows (Brandt 1987):

- Natural soil development in humid regions includes base leaching and acidification. Acidic precipitation can intensify these processes;
- The extent to which soils can be acidified depends on the initial pH and buffering capacity of the soils. Soils naturally low in basic cations, with low pH and limited cation exchange capacity, may be particularly sensitive;
- Soil acidification is detrimental to many microbiolgical processes in soils, for example the mineralization and humification of organic matter and the mobilization and fixation of nutrients and toxic substances (e.g., toxic metallic ion concentrations);
- Use of fertilizers, especially nitrogen fertilizers with physiological acidic reactions, as well as other cultivation and land-use practices can increase soil acidification.

The soil factors most likely to be affected by acidic deposition are (1) an increase in solubility of toxic metals such as aluminum and manganese so that they affect roots or mycorrhizae; (2) an alteration of nutrient supply through leaching or by altering microbial populations or processes (toxicity of mycorrhizae due to increased nitrogen has been postulated); and (3) a reduction in the solubility of phosphorus and molybdenum. Acidic deposition can increase natural acidification processes; however, at current levels the contributions are considered to be minor (Altshuller and Linthurst 1984; Binkley and others 1987). The likelihood of increased acidification of soils occurring is discussed in the paragraphs that follow.

Soil acidification is the primary condition controlling metal mobilization. Acid soils often contain increased concentrations of aluminum in the soil solution. A substantial body of knowledge has accumulated over the years concerning the toxicity of aluminum to agricultural crops. Many plants are unable to tolerate high aluminum concentrations because they impair root activity. Excess aluminum in highly acid soils causes root necrosis and reduced root growth; these in turn increase plant susceptibility to moisture and/or nutrient stress (Black 1968; Foy and others 1978; Schier 1985). Many tree species, however, grow on very acidic soils and appear to tolerate the high aluminum concentrations in soil solutions. Forest soils are commonly acid with pH values of 3.5 to 5.5. Tree species may become intolerant if high aluminum to nutrient ratios limit root uptake of calcium and magnesium and create a nutrient deficiency (Binkley and others 1989).

Shortle and Smith (1988) attributed aluminum-induced calcium deficiency syndrome in declining red spruce to restricted calcium uptake by trees growing in highly acid soils. Though soluble aluminum in high concentrations is toxic to the roots of a wide range of agricultural and forest plants, there are healthy stands of trees on soils of similar acidity and with similar aluminum concentrations both in the soil and fine roots. In addition to toxicity, aluminum interferes with calcium uptake by fine roots when the soluble ions are in equimolar concentrations. Shortle and Smith propose that inhibition of calcium uptake causes decline in cambial growth and a loss of crown in mature red spruce.

The demand for calcium per unit area of cambium surface is essentially constant and the surface area expands exponentially as mature trees add secondary xylem. Restricted uptake of calcium suppresses cambial growth, and a decrease in the widths of annual rings results. This reduces the amount of functioning sapwood since the older sapwood is continuously transformed into heartwood. Both declining and healthy trees have the same number of annual rings, but in healthy trees the rings are wider. Trees become susceptible to death from secondary pathogens and insects when the amount of sapwood forms less than 25 percent of the cross-sectional area. Crown density and associated leaf mass and area are positively correlated with the cross-sectional area of sapwood. As the cross-sectional area of sapwood decreases, crown density also would be expected to decrease (Shortle and Smith 1988).

A comparison by Shortle and Smith (1988) of trees growing on Mt. Abraham, VT, where more than half the canopy spruce are dead, with trees growing near Beddington, ME, where canopy spruce are uniformly healthy, supports their theory of aluminum-to-calcium imbalance. The humus layer for all stands

investigated had a pH of 3.2 to 4.6. Equilibrium soil solution analysis indicated that the aluminum-to-calcium ratio was greater than 1 on Mt. Abraham and was significantly higher than in the soil solution from Beddington. Spruce trees at high elevations on Mt. Abraham (1000 m) have been most affected because the aluminum-to-calcium ratio is greater than 1 in both humus and subsoil, and limits calcium supply. Continued input of sulfur and nitrogen anions will increase the amounts of aluminum in solution, and the increased aluminum will reduce calcium uptake. Aluminum toxicity is not the problem under these circumstances (Shortle and Smith 1988).

At present it is not known whether acidic deposition is inducing nutrient leaching in the forest soils of eastern North America because: a) the nutrient status of eastern forests is not characterized well enough to make such a determination, and b) in cases where nutrient leaching is occurring, it is not known whether the amounts being leached are greater than would be expected in the absence of ambient acidic deposition. Turner and others (1986) estimated that base-cation leaching would result in growth reductions in 51 percent of forested soils in the Southeast. The Piedmont and Coastal Plain were judged to have the largest proportion of areas sensitive to acidic deposition (Turner and others 1986). Some have questioned the data used to establish sensitivity by Turner and associates. Binkley and others (1989), however, state that the data base used by Turner and co-workers is probably the "most extensive and quality-assured" available.

Turner and others (1986) estimated that it would take approximately 30 years at current rates of acidic deposition to decrease soil base saturation of the most sensitive forest soils from 60 to 40 percent, and 80 years from 60 to 25 percent. Replenishment of base cations by deposition was not considered.

Tabatabai (1985) predicts that at current rates nutrient leaching induced by atmospheric deposition is unlikely in most eastern soils because: (1) sulfate adsorption capacities are relatively large; (2) cation exchange capacities are large; (3) most forests are nitrogen limited and most nitrate will be taken up by forest vegetation; and (4) soil base cations are being replenished by atmospheric deposition and mineral weathering.

Field studies of base cation and anion leaching in eastern forests indicate that the amounts of these ions are insignificant in soil leachate of most forest watersheds studied (Walker Branch and Camp Branch, TN; Hubbard Brook, NH; and Huntington Forest, NY) (Bormann 1979; Johnson 1983; Johnson and others 1985a; Johnson and others 1985b; Mollitor and Raynal 1982).

Binkley and others (1989) report that to date, forests in the South have responded most strongly to additions of phosphorus and nitrogen, probably because growth of most forest stands in the South is phosphorus and nitrogen limited. There is, however, some evidence that deficiencies in other elements have also limited growth. Data on phosphate and molybdenum availability in acidified soils are limited; however, molybdenum deficiency in southern forests has not been reported. Potassium deficiencies may be common on sandy soils.

Though soil acidification does affect many biological processes, at present it is difficult to distinguish between direct effects of acidification and secondary effects resulting from acid-induced changes in the soil solution. Important effects under field conditions have not been demonstrated (Altshuller and Linthurst 1984). The possible effects of nitrogen toxicity to mycorrhizae have been reviewed (Cline and others 1987). The authors concluded that at the current rate of nitrogen deposition in the southeastern United States, nitrogen is not toxic to mycorrhizae.

Summary.--The consensus of most investigators at the present time is that no apparent drastic or potentially catastrophic short-term effects result from the atmospheric deposition of gaseous air pollutant chemicals on soils. In general, the long-term effects are unknown. Concern exists chiefly for the possible long-term effects of acidic deposition. Most studies have emphasized changes in soil pH resulting from deposition of acidic or acidifying substances. Deposition of sulfate and nitrate may pose a greater problem.

Data suggest that nitrogen in excess of the amounts required for growth of trees is accumulating in coniferous forests. Appearance of nitrate in the soil solution and in the xylem sap of conifers are symptoms of this problem. Nitrate appears in the xylem sap because the roots of densely growing conifers are unable to metabolize large amounts. In addition, excess nitrogen in foliage of boreal and subalpine conifers does not enhance photosynthesis. It not only alters the manner in which carbon is allocated between shoots and roots but also provides nitrogen in a form difficult to metabolize and one that increases sensitivity to frost. Resulting nutrient imbalances are likely to make conifers more susceptible to drought, other pollutants, and pathogens. Competition under these circumstances favors deciduous species.

Soil acidification is the primary condition controlling metal mobilization. An increase in the concentrations of metals such as aluminum and manganese toxic to roots and mycorrhizae of has been postulated as a possible result of acidic deposition. While many plants are sensitive to high aluminum concentrations in soil solutions, healthy stands of trees do grow on very acidic soils (pH 3.5 to 5.5) and appear to tolerate the high aluminum concentrations. Aluminum, on the other hand, also interferes with calcium uptake by fine roots when soluble ions are in equimolar concentrations. Calcium deficiency results in reduced cambial growth, reduced formation of new sapwood, and a decrease in functioning sapwood as the older sapwood is transformed into heartwood. Both declining and healthy trees have the same number of annual rings, but in healthy trees the rings are wider. When sapwood forms less than 25 percent of the cross-sectional area, trees become susceptible to death from secondary pathogens and insects.

At the present time the effects of acidic deposition on nutrient leaching are not known. Tabatabai (1985) predicts that at the current rates atmospheric deposition is unlikely to induce nutrient leaching in most eastern soils. Field studies of base cation and anion leaching in eastern forest suggest that the amounts of these ions in soil leachates for most forest watersheds are insignificant.

Current data suggest that the forests in the South have responded most strongly to additions of phosphorus and nitrogen. There is, however, some evidence that other nutrients have also limited growth. Aqueous aluminum may

limit the availability of phosphorus and molybdenum; however, there is no evidence of reduced molybdenum availability. Potassium may be deficient in sandy soils (Binkley and others, 1989).

Though soil acidification affects many biological processes, important effects under field conditions have not been demonstrated. Under current rates of nitrogen deposition in the Southeastern United States, nitrogen is not toxic to mycorrhizae.

4.5 What Effects are Airborne Chemical Pollutant Stresses Having on the Forest Ecosystems of Eastern North America?

Forest decline is reported to be an established fact in eastern North America (Johnson 1985; Klein 1985; Klein and Perkins 1987; Tomlinson 1983). Changes in growth and decline and mortality of certain tree species have been reported for high-elevation forest ecosystems from Maine, New Hampshire, Vermont, and New York, south to North Carolina and Tennessee. Studies indicate that the decrease in growth of forest trees began during the late 1950's or early 1960's (Adams and others 1985; Benoit and others 1982; Johnson and others 1984; McLaughlin and others 1982; Phipps and Whiton 1988). The extent of the forest declines and the factors that precipitated them, however, are questions of controversy (Klein and Perkins 1987; Taylor and Norby 1985). The problem is establishing causation. Woodman and Cowling (1987) point out the need for rigorous proof of cause; however, only circumstantial evidence is available because growth reductions began at least 30 years ago. Associating the growth reductions with air pollution injury is even more difficult, because data indicating the presence of concentrations of phytotoxic air pollutants are not available. Limited monitoring of SO_2 and NO_2 did not begin until 1959 or 1960. Though the Continuous Air Monitoring Project (CAMP) came into existence in 1962, valid oxidant data for the eastern United States were not available until 1964, and then only for the cities of Chicago, Cincinnati, Denver, Philadelphia, St. Louis, and Washington, DC (U.S. Department of Health Education and Welfare 1968).

Despite the absence of data measuring specific concentrations of SO_2, NO_2, and O_3, emissions trends data for SO_x and NO_x indicate a sharp overall increase beginning in the late 1940's (see figs. 5 through 9). Increases in ozone concentrations undoubtedly also began during the late 1940's soon after World War II when the large-scale emissions of NO_x began. Nitrogen oxides are intimately associated with formation of ozone via the photochemical oxidant cycle. Unquestionably, increasing NO_x emissions fueled the formation of ozone even though data do not exist to indicate this.

An additional factor that makes causation difficult in determining the effects of pollutant stress on ecosystems is that mature forest ecosystems, as mentioned at the beginning of this chapter, are not completely stable, but maintain themselves in an oscillating steady state (Kozlowski 1985). Severe competition among trees and understory vegetation for light, water, and mineral nutrients results in the elimination of old and suppressed trees and the addition of new ones. The elimination of those trees most susceptible to competitional stresses is accelerated by additional and severe periodic stresses such as drought, flooding, fire and attacks by insects or disease-causing organisms. Succession results (Kozlowski 1985). Pollutant stresses,

which can be either severe episodic stresses (1 day to a week of acute high exposures) or long-term chronic stresses, are superimposed upon the naturally occurring stresses already being encountered by the trees. They may accelerate the processes of change already under way. The effects of the natural stresses upon ecosystems, unless they are catastrophic (e.g., fire, flood, or windstorm), are frequently difficult to determine.

Causation for ecosystem breakdown in eastern North America has been definitely established only around point sources of pollution. Emissions of SO_2 from smelters and an iron sintering plant resulted in trees and shrubs being eliminated from the vicinity of Copper Hill Basin near Ducktown, TN (Hedgcock 1914), and from around Wawa and Sudbury, Ontario, Canada (Gordon and Gorham 1963; Linzon 1978). The pollutant dosage in the above situations was catastrophic, resulting in long-term injury to the ecosystems.

Responses of forest communities or ecosystems to catastrophic or acute disturbances are, as a rule, readily observable and measurable. Catastrophic disturbances cause mature forests to be returned to an earlier successional stage. During normal succession in unpolluted atmospheres, the number of species in a community usually increases. Productivity, biomass, community height, and structural complexity also increase. Stress alters succession and diverts energy from growth and reproduction to maintenance. Biomass accumulation and production decrease and there is a reduction in structural complexity, species diversity, environmental modification, and nutrient control. Succession reverts to an earlier stage.

The responses of forest ecosystems to air pollution are integrated responses because the air pollution stress is superimposed upon and interacts with the multiple stresses the component species are already experiencing. The composition and successional stage of forests are determined by the kind, frequency, and magnitude of disturbance (table 16) (Kozlowski 1985).

Studies of radiation effects (Woodwell 1970) and data from air pollution effects around strong point sources (see above), as well as studies and observations of ozone effects on natural ecosystems such as the San Bernardino Forest, suggest that forest ecosystems respond to increasing air pollution stresses in a predictable pattern that may be thought of as a continuum of responses (table 16) (Bormann 1985; Guderian and Kueppers 1980; Kozlowski 1985; Smith 1981; Woodwell 1970). Circumstantial evidence for forest change may be obtained by comparing field data gleaned from studies of ambient pollution effects on individual trees, surveys to determine visible injury symptoms on forest trees and vegetation in the field, and studies of soil changes with the continuum of responses (table 16) of Bormann (1985).

The responses of decline and dieback seen at the present time in eastern North America are the result of chronic stresses. Responses of forest ecosystems to chronic stresses are diffuse and, therefore, less readily observed and measured. Subtle and indirect effects of pollutant dosages on individual trees and other perennial species are cumulative. Sensitive annual species respond during the year they are exposed. Cumulative pollutant stress provides a selective force that eliminates those species that lack the genetic diversity to survive, suppresses some genotypes, and favors others. The distribution of a species and the productivity of an ecosystem both depend upon the success or

failure of individual plants (Billings 1978). When removal of an individual species alters species interactions and changes population dynamics, then energy flow and chemical nutrient cycling of a community or ecosystem also are altered, thus setting the stage for changes in community structure that possibly could be irreversible (table 16) (Guderian and Kueppers 1980). The problem, once the changes have occurred or are occurring, is to discover what caused the changes. Because of lack of data (monitoring data in particular), we are left with hypotheses (table 17).

Surveys made in 1982 give quantitative evidence of spruce decline in stands throughout the Appalachian Mountains from New Hampshire and Vermont to North Carolina (Johnson and Siccama 1983). Red spruce (Picea rubens Sarg.) is the most characteristic species of the subalpine coniferous forests that occupy the higher peaks and ridges of the Appalachian Mountains from Maine to North Carolina and Tennessee. In the North, balsam fir [(Abies balsamea (L.) Mill.)] is commonly present as a codominant species, whereas Fraser fir [(A. fraseri (Pursh) Poir.)], a closely related species, is the codominant in the South (Adams and others 1985).

In the high elevation forests of New York, Vermont, and New Hampshire, red spruce has shown marked dieback and a large reduction in basal area and density. Spruce mortality has occurred rather evenly regardless of tree size, variety of stand composition, and growth on a variety of soils. Red spruce dieback was observed in substantial areas of Vermont and New Hampshire in the 1950's and 1960's. The areas were studied to determine the cause. No primary pathogens or insect infestation were observed on the trees. Fungal pathogens present (Polyporus borealis, Fomes pini, and Armillaria mellea) were those whose invasion is usually triggered by predisposing agents. Data from long-term (1780 to 1965) tree ring records indicated large fluctuations in annual growth, but no consistent pattern of declining growth (Johnson and Siccama 1983).

The red spruce dieback is characteristic of a stress-related disease. These characteristics include the absence of an obvious cause, dieback (loss of foliage beginning in the crown and progressing over time downward and from branch tips inward), reduced vigor, and subsequent invasion by secondary organisms that normally do not cause substantial injury to vigorous trees (see table 16, II-3) (Johnson and Siccama 1983; Siccama and others 1982). Spruce regeneration is reported to be severely depressed.

Hardwood forests also show quantitative decline. There is a reduction in mosses and mycorrhizae, and the natural litter decomposition process has been affected. Seedlings have malformed roots, and in the spring when plant growth is most vigorous, the highly acidic soil nutrient solution contains toxic concentrations of metals (Klein 1985). Many hypotheses have been suggested to explain these effects (table 17). The multiple stress hypothesis is most current (Johnson and Siccama 1983, Klein and Perkins 1987).

In the Southeast, the decline and mortality of the Fraser fir in the Great Smoky Mountains National Park, Plott Balsam Mountains, NC, and the Black Mountains, which include Mt. Mitchell, have been attributed to infestation by the balsam woolly adelgid (Hain and Arthur 1985). Results from a 20-year experimental study of Fraser fir growing in the Smoky Mountains

Table 16. Ecosystem Response to Pollutant Stress

Continuum	Stage of Response of Vegetation	Response of Ecosystem
0	Anthropogenic pollutants insignificant	Unaffected; systems pristine
I	Pollutant concentrations low; no measurable physiological response	Ecosystem functions unaffected; pollutants transferred from atmosphere to organic or available nutrient compartments
II	Pollutant concentrations injurious to sensitive species (1) Reduced photosynthesis, altered carbon allocation, reduced growth and vigor (2) Reduced reproduction (3) Predisposition to entomological or microbiological stress	Altered species composition; populations of sensitive species decline; some individuals are lost. Their effectiveness as functional ecosystem members diminishes; they could be lost from the system. Ecosystem reverts to an earlier stage.
III	Severe pollution stress. Large plants of sensitive species die. Forest layers are peeled off: first trees, tall shrubs and, under most severe conditions, short shrubs and herbs.	(1) Simplification, basic ecosystem structure changes, becomes dominated by weedy species not previously present. (2) Reduced stability and productivity; loss of capability for repairing itself. Runoff increases, nutrient loss, and erosion accelerates; a barren zone results. Ecosystem collapses.

Source: Adapted from Bormann (1985); Kozlowski (1985); Smith (1974);

National Park indicate that the balsam woolly adelgid had killed almost all of the canopy trees and had reduced basal area in two plots established in the 1960's. Spruce basal area remained about the same for the same period. The report does not mention monitoring of atmospheric pollutants or possible pollutant-insect interaction or predisposition (Busing and others 1988). Recent studies on Mt. Mitchell, however, do not attribute the death of Fraser fir solely to the balsam woolly adelgid, but suggest that atmospheric deposition and multiple pollutant stresses also have a role in tree mortality (Bruck and others 1988; Hain and Arthur 1985). The studies also cited exposure to gaseous air pollutants, particularly ozone, and cloud water deposition of acidic substances as among the possible stresses having increased host susceptibility to attack by the balsam woolly adelgid (Bruck and others 1988; Hain and Arthur 1985).

Table 17. Some Hypotheses to Explain Forest Decline in Eastern North America

Category	Hypothesis
Natural process	Synchronous canopy dieback Biotic pathogens Assorted abiotic stress agents
Forest stand history	Landscape conversion Forest management practices (e.g., genetic stock) Forest fire frequency and intensity
Anthropogenic stress	Nitrogen subsidy (e.g., nitric acid vapor, nitrate deposition) Direct ozone effects (e.g., inhibition of photosynthesis) Indirect ozone effects (e.g., leaching) Direct effects of hydrogen ion deposition Indirect effect of hydrogen ion deposition (e.g., rhizosphere aluminum toxicity) Deposition of sulphur Deposition of organics Deposition of multiple pollutants (wet and dry deposition) Deposition of trace elements Acid cloud moisture
Multiple factors	Site-specific combination of assorted natural processes, forest stand history, and anthropogenic stress

Adapted from Taylor and Norby (1985).

Soil conditions resulting from the deposition of airborne sulfur- and nitrogen-derived chemicals have been hypothesized as having been involved in the dieback and decline of red spruce and responsible for the slowdown in growth of other trees (sec. 4.4). The deposition of nitrogen in excess of amounts required for coniferous tree growth as well as the imbalance of aluminum and calcium in the fine roots of trees have recently been proposed as the cause. Nitrogen in excess of growth requirements can be detrimental to conifers. In eight regions where forest decline has been observed, the nitrate concentration in the soil was an order of magnitude higher than in areas where forest decline has not yet been observed (Waring 1987). Enrichment of the soil with atmospheric nitrogen in excess of growth requirements of boreal and subalpine conifers will not necessarily enhance growth, but is likely to cause nutrient imbalances and make conifers more susceptible to drought, other pollutants, and pathogens (Waring 1987). An imbalance of calcium and aluminum in the fine roots of trees reduces the rate of wood formation, decreases the amount of functional sapwood and live crown, and results in large trees being more vulnerable to secondary diseases and insect pests (Shortle and Smith 1988).

In the preceding sections (4.2.1 to 4.2.7), the authors have discussed both field observations and experimental data dealing with the effects of acidic deposition, sulfur dioxide, nitrogen oxides, ozone, and gaseous pollutant mixtures on individual trees and seedlings via foliage-mediated response mechanisms. Exposure of trees to sulfur dioxide and ozone in their natural habitats has been shown to inhibit photosynthesis; alter carbon allocation; decrease vigor; disrupt reproduction; change host susceptibility to insects, pathogens, and mycorrhizal colonization; and, in the final analysis, affect tree growth. Evidence that nitrogen oxides or gaseous pollutant mixtures cause growth reductions under ambient conditions is lacking.

Sulfur dioxide is emitted from point sources. In the eastern United States, 70 percent of the emissions are from power plants with tall stacks. In the summer, residence time in the Midwestern States is approximately 2 days. During that time, the plumes from the power plants are likely to have been transported up to 1000 km, broadened, and the SO_2 to have been transformed into sulfate or to have been removed by dry or wet deposition (Husar and Holloway 1983; sec. 3.4). The highest average SO_2 concentrations at rural sites in eastern North America are in the Ohio River Valley with values of 10 ppb. Average concentrations decrease with increasing distance from this area, reaching approximately 2 ppb in New England and 1 ppb in the South (Mueller and Hidy 1983).

Ozone, on the other hand, is a regional gaseous air pollutant (see chapter 3.). It affects vegetation throughout the United States, impairing crops, native vegetation, and ecosystems more than any other air pollutant (Daines and others 1960; Heck and others 1980; U.S. Environmental Protection Agency 1986). Ozone is episodic and may be transferred long distances. During an episode, ozone trajectories may cover very large areas. For example, an ozone episode covering most of a 20-state area occurred April 12-23, 1976 (Wolff and others 1977). During this episode, ozone concentrations exceeding 80 ppb occurred simultaneously across the Midwest and the Northeastern United States. On the afternoon of April 12, ozone concentrations in excess of 80 ppb were measured in Indiana and Ohio ahead of the high pressure system that moved out of Canada and was centered over Lake Michigan. By the evening of April 13, the high ozone concentrations (60 to 80 ppb) observed over Indiana and Ohio were advected to the East Coast of the Mid-Atlantic States, with trajectories extending from Ohio to New Jersey. The center of the high pressure system continued to move southeastward, and by the morning of April 16 was centered off the coast of North Carolina. At this time, concentrations ranging from 60 to 80 ppb extended from Maine to Virginia and from Massachusetts to Virginia in the Boston to Washington corridor (Wolff and others 1977).

Additional studies indicate that two other ozone episodes that took place during August 1-6 and August 8-14, 1976, exhibited trajectories similar to the one described above (Wolff and others 1980). In both cases, the high pressure originated in Canada and moved southeastward into the Midwest. Concentrations ranged from 30 to 50 ppb in the high as it moved into the U.S. During the first or second day that the center of the high was over the Midwest, two areas (the Midwest and the Northeast Corridor) developed ozone concentrations greater than 80 ppb. One area extended from the western shore of Lake Michigan to St. Louis, across eastern Indiana, Ohio, southeastern Michigan, and

northwestern Pennsylvania to southern Ontario. The second area was the Washington, D.C.-Boston, MA, corridor. As the high pressure area persisted, the two areas merged and the ozone concentrations continued to rise until a cold front moved down from Canada. Maximum ozone concentrations observed during the two August episodes exceeded 200 ppb (Wolff and others 1980). The second episode varied from the first in that the trajectory extended south over Lexington, KY. In both studies, the episodes included the area from West Virginia and Virginia in the South to Maine in the Northeast. Ozone concentrations in trajectories increase when the air parcels pass over urban areas where additional ozone is present. For example, in the above episode, an air parcel bearing ozone concentrations exceeding 100 ppb added another 100 ppb when passing over Philadelphia (Wolff and others 1980). The above studies point out that ozone episodes are not an abnormal phenomenon, but can occur several times during a single year.

All ozone episodes are not associated with high pressure systems originating in Canada. Studies of ozone episodes made during July 12-21, July 21-24 and July 26-30, 1977, indicate that ozone from the western coast of the Gulf of Mexico also may be transported into the Northeast (Wolff and Lioy 1980). During the episode of July 12-21, 1977, ozone transport was associated with a high pressure area over the Gulf of Mexico. Air parcels originating in the vicinity of Texas and Louisiana traveled northeastward through the Midwest to the Atlantic Coast. The episode that began on July 12 lasted until July 21, with the area extending from the Texas-Louisiana Gulf Coast to the northeastern Atlantic Coast being exposed to ozone concentrations averaging 120 to 130 ppb. The highest concentration of 328 ppb was observed in Connecticut (Wolff and Lioy 1980). Unlike the July 12-21 episode, the second and third episodes were associated with high pressure systems that moved down from Canada and suppressed the tropical gulf air. The ozone "river" reaching from the Gulf Coast to New England that had come into existence during the July 12-21 episode was prolonged when the cold fronts of July 21 and July 23, 1977, from Canada moved eastward and off the Atlantic Coast. These episodes simultaneously exposed nearly two-thirds of the United States, an area 2×10^6 mi^2 (Wolff and Lioy 1980).

The foregoing discussion not only depicts the episodic nature of ozone exposures, but also points out the fact that a major portion of the United States east of the Mississippi River has for many years been frequently exposed to phytotoxic ozone concentrations. Even though the understanding that ozone was not just a local problem but could be transported long distances did not develop until during the 1970's, ozone episodes had been occurring for some time. Linzon (1960) points out that by 1957, foliar injury of white pine had been reported (14 references are cited by Linzon) throughout its range under a variety of names, though the cause of the injury was still unknown. Foliar injury of white pine under the name of "white pine needle blight" was reported at the Petawawa Forest Experiment Station, Chalk River, Ontario, during the month of July in 1957 and in July and September 1958. During 6 of the 8 years from 1957-1964, foliar injury was observed on white pine. Ozone was suspected as a cause of the injury, but attempts to establish a definite relationship were not successful though ozone concentrations sufficient to cause "weatherfleck" of tobacco were observed (Linzon 1967).

In the southeastern United States, foliar injury of eastern white pine resulting from ozone exposure also has been observed for some time. Berry and Ripperton (1963) reported that, though intensive studies to determine the cause of "emergence tipburn" of eastern white pine were not begun in Pocahontas County, WV, until 1957, many people in the same area had been reporting casual observations regarding the phenomenon for over 20 years. We now know "emergence tipburn" and "white pine needle blight" to be visible symptoms of ozone injury. Berry (1961) reported that "emergence tipburn seems to be more prevalent and more damaging in the northerly latitudes, being very common in the Northeastern United States and Canada, and rare on white pine in Georgia." Further, Berry cites the occurrence of an ozone episode during July of 1961 when he states that an unusually high incidence of "emergence tipburn" occurred in western North Carolina and "surrounding States." In other words, ozone episodes were occurring long before scientists became aware, not only of their existence, but of the fact that ozone episodes were causing the injury observed on white pine. Hence, beginning no later than the early 1950's, both natural and agricultural ecosystems from the Gulf Coasts of Texas and Louisiana to the Midwest and Canada and throughout the East from North Carolina and Tennessee to Maine have been exposed for many years. Hayes and Skelly (1977) associated injury to eastern white pine at three rural sites in Virginia between July 1-5, 1975, with a high pressure system over the Great Lakes and a low, Hurricane Amy, off the Atlantic Coast. Air parcels bearing ozone moved from the Northeast and Midwest into Virginia. The episode was dissipated when the cold front from the Midwest moved across Virginia into the Atlantic Ocean. Since this time, when ozone episodes have occurred in the same region, they have been associated with meteorological phenomena such as cited above (Skelly and others 1984).

Vegetation injury due to ozone was not unknown in the eastern United States in the 1940's, though it was not identified as such until 1960. An unusual pattern of foliar injury of many cultivated crops, ornamentals, and indigenous vegetation was observed in the New Jersey area beginning in 1944. The injury was frequently followed by partial or complete defoliation of many deciduous and coniferous plants (Daines and others 1960). Ozone was first recognized as the causal factor of the foliar injury after Heggestad and Middleton (1959) recognized that weatherfleck of tobacco was due to ozone injury. Their observations indicate that during an ozone episode in October 1958, concentrations at Beltsville, MD as measured with a Mast meter reached 380 ppb on October 9, 500 ppb on October 10, and 430 ppb on October 16, 1958, respectively. Ozone injury to crops in New Jersey during the 1959 growing season, reported by Daines and others (1960), indicates that another episode occurred that year during the growing season.

It is possible, therefore, that the growth reductions in forest trees observable today and attributed to many different factors (table 17) are the cumulative responses from exposure to multiple O_3 episodes that began many years ago.

Many reports point out that trees at high altitudes frequently are observed to more severely injured than those lower on the mountain. This could be due to O_3 exposure because the diurnal behavior of ozone at higher altitudes results in high altitude forest ecosystems suffering greater ozone injury than those at lower altitudes (Lefohn and Jones 1986; Wolff and others

1987). Though daily maximum and midday ozone concentrations are similar at different altitudes, the dosage increases with height. Ozone is rapidly depleted near the surface below the nocturnal inversion layer. However, mountainous sites above the nocturnal inversion layer do not experience this depletion. Therefore, the total exposure to ozone in mountainous areas can be much higher than in nearby valleys (see table 18). Because of this, several considerations need to be taken into account when assessing exposure-response relationships of forest ecosystems at high altitudes:

1. Sites at elevations above the nocturnal inversion layer can be exposed to higher peak and higher total concentrations of ozone than sites at lower elevations (Miller and others 1977; Miller and others 1982; Wolff and others 1987).

2. The maximum ozone concentrations observed at elevated mountainous sites, as well as at many nonmountainous rural and remote sites, often occur at night (Lefohn and Jones 1986; Wolff and others 1987). For species such as white pine, in which the stomata remain fully or partially open after dark, this is particularly important.

3. Sites at higher elevations are often exposed to sustained or multiple peak concentrations of ozone within a given 24-hour period as the result of conditions such as (a) trapping inversions; (b) the successive transport of ozone from multiple urban sources upwind, either aloft or across terrain devoid of sufficient ozone scavengers; and (c) the occurrence of mountain-valley and upslope-downslope flows, such that the trajectory of an air parcel passes back over the same forest or stand of trees (Wolff and others 1987).

Table 18. Comparison of Ozone Data at Different Altitudes and at an Urban Area

	7/22/75		7/23/75		
Location	1-hr max ppb	24-hr mean ppb	1-hr max ppb	24-hr mean ppb	No. of hr ≥80 ppb
High Point, 500 m	66	49	130	81	13
High Point, 300 m	61	38	110	61	9
High Point, 140 m	59	26	120	52	9
Bayonne, NJ, sea level	69	33	119	48	7

High Point Mountain is in New Jersey.

After Wolff and others (1987).

Most studies of forest ecosystems have monitored only the daylight ozone concentrations. Thus, the reported exposures may not represent either the number length or magnitude to which the forests have been exposed (U.S. Environmental Protection Agency 1986).

The chronic nature of the episodic ozone stresses to which the ecosystems of eastern North America have been subjected has made it difficult to determine what changes, if any, have resulted from them. Some, but not all, eastern white pine trees are sensitive to ozone. McLaughlin and others (1982) and Benoit and others (1982) divided the species into three categories: sensitive, intermediate, and tolerant. The sensitive members of the white pine species are indicators that a forest stand, community, or ecosystem has been exposed to ozone, but do not necessarily inform scientists whether the ecosystem as a whole is being affected. Many tree species are sensitive to ozone. Davis and Wilhour (1976) used foliar injury to determine ozone sensitivity. Twenty-three species of trees grown in North America were listed as sensitive and 18 as intermediate. More recently, Pye (1988), using experimental data on seedlings, stated that ozone can reduce photosynthesis and growth of 43 tree species and hybrids.

Given the fact that ozone has been a regional stress for decades, the most sensitive plant species have undoubtedly been removed and supplanted by tolerant species. Hayes and Skelly (1977) reported that in 1971, 15 sensitive white pine trees were tagged. After 2 years of exposure to ambient ozone concentrations, 13 had declined in health, and after 3 years of consecutive ozone exposure, 6 of those 13 were dead. The possibility that some plant species may have evolved genetic resistance has been proposed (Taylor and Norby 1985).

Selection for genetic resistance has undoubtedly occurred in urban habitats. As with many agricultural crops and some forest tree species, trees and other woody plants that are grown in a variety of urban habitats have been selected over time for viability in stressed environments. Umbach and Davis (1984) point out that trees obtained from commercial nurseries are not likely to represent the full range of genotypes present in the wild population of the species. The work of Rhoads and others (1980), along with that of Karnosky (1981, 1983), suggests that, based on foliar injury, the majority of the plants growing along streets and urban parks, arboreta, remnant woodlots, and suburban communities are relatively insensitive to O_3 exposure. Their relative insensitivity may be the combined result of genetic selection and physical factors affecting stomatal processes.

Unlike the ponderosa or Jeffrey pines in the San Bernardino Forest, the eastern white pine is not what has recently been termed a "keystone species," one so important to its ecosystem that its removal or demise will cause the collapse of that ecosystem (Sunquist 1988). Therefore, the demise of the white pine, as in the situation cited above, did not make a readily observable impact on the ecosystem. Except for the observations of McClenahen (1978) on the subtle changes occurring in forests when exposed to pollutants for 40 years, and the experiments of Duchelle and others (1982) to determine the effect of ozone exposure on native vegetation in the Blue Ridge, no studies have been made in the eastern United States to determine how exposure to ozone has affected ecosystem components other than trees.

Had the deaths of sensitive eastern white pine resulted in dieback and mortality as severe as have the deaths of the codominant spruce and fir trees in the high elevation forests of North Carolina, New Hampshire, and Vermont, our knowledge concerning the impact of ozone on the forest ecosystems of

eastern North America would be much greater today. As it is, because we lack sufficient data, we can only speculate about the changes that undoubtedly have resulted from long-term ozone exposure.

Summary--The forest ecosystems of eastern North America are in the process of change. Decreases in growth, dieback, and mortality of certain tree species have been reported in the high-elevation forests ecosystems from Maine, New Hamsphire, and Vermont south to North Carolina and Tennessee. Dendroecological studies indicate that the decrease in growth of forest trees began during the 1950's or early 1960's. The extent of forest declines and the factors that precipitated them, however, are questions of controversy. The problem is establishing causation. Many hypotheses have been advanced as to the possible cause or causes.

Attributing the observed growth reductions to air pollution is difficult because data indicating the presence many years ago of injurious concentrations of phytotoxic air pollutants is lacking. Of the gaseous air pollutants, ozone is the most phytotoxic. Sulfur dioxide injury to vegetation is found most frequently in the vicinity of strong point sources. Vegetation exposure to phytotoxic concentrations in these areas is usually of short duration. (Exceptions are Wawa and Sudbury, Ontario, Trail British Columbia, and Copper Hill Basin near Ducktown, TN, where catastrophic emissions resulted in long-term injury to the ecosystems). Nitrogen oxides have not been measured in the atmosphere in concentrations sufficient to cause vegetational injury.

Ozone is a regional pollutant and is, therefore, the only one capable of having exposed the entire area where vegetation injury and growth reductions have been observed over the past several years without leaving a trace. Ozone molecules are ephemeral. By-products of ozone, unlike residuals of sulfur or nitrogen oxides, fluorides, heavy metals, or radionuclides, are not selectively retained or accumulated in any particular components of the ecosystem.

Limited monitoring of SO_2 and NO_x concentrations in the Eastern United States did not begin until 1959 or 1960, while valid oxidant data did not become available until 1964 and then only for certain large cities. However, emission trends data indicate a sharp increase for SO_x and NO_x beginning in the late 1940's. Increase in ozone concentrations, fueled by large-scale NO_x emissions that began during World War II, probably started in the late 1940's even though data do not exist to indicate this. Circumstantial evidence suggests that the ecosystems of eastern North American have been subjected to episodic phytotoxic concentrations of ozone similar to those cited below for at least 30, possibly 40 years.

Typical ozone episodes in the Eastern United States are associated with high-pressure systems which originate out of Canada and move southeastward into the Midwest enroute to the Atlantic Coast. Ozone trajectories during an episode may cover very large areas. For example, an episode covering most of a 20-state area occurred April 12-23, 1976. During this episode, ozone concentrations exceeding 80 ppb occurred simultaneously from the Midwest across to the Atlantic Coast and into the northeast. Additional studies indicate that two other ozone episodes, each lasting one week and exhibiting trajectories similar to the one mentioned above, took place during August, 1976. Maximum concentrations measured during the two August episodes were 200 ppb.

The following year there were three episodes, July 12-21; July 21-24 and July 25-30, 1977. The last two, like the 1976 episodes, were associated with high-pressure systems originating in Canada, but the first episode originated in the Texas-Louisiana area and formed "a river of ozone" from the Texas Gulf Coast through the Midwest to the Atlantic Coast. The highest concentration of 328 ppb was observed in Connecticut. Ozone concentrations in trajectories increase as air parcels pass over urban areas where additional ozone is present. There also have been reports for many years of ozone episodes originating in the Washington-Philadelphia area and moving westward and south across West Virginia and Virginia down into the Carolinas.

Published reports of field observations support the view that ozone episodes have been occurring for at least 40 years. Vegetation injury in the eastern United States resulting from ozone was first observed in the 1940's, though it was not recognized as such until 1960. Beginning in 1944, an unusual pattern of foliar injury of many cultivated crops, ornamentals, and indigenous vegetation was observed in the New Jersey area. Injury was frequently followed by partial or complete defoliation of many deciduous and coniferous species. Ozone was first recognized as the causal factor after Heggestad and Middleton in 1960 reported that weatherfleck of tobacco observed during the 1958 growing season in Beltsville, MD was due to ozone. Ozone injury of crops in New Jersey also was observed during the 1959 growing season.

Field observations also suggest that oxidant-induced injury of vegetation has been occurring in the Appalachian Mountains for many years. Studies of forest decline of eastern white pine in an area covering several hundred square miles of the Cumberland Plateau of East Tennessee in the 1950's concluded that unidentified atmospheric constituents were the cause. In addition, people living in Pocahontas County, WV in 1957 had been reporting casual observations of visible ozone injury of eastern white pine for more than 20 years.

Forest ecosystems at high altitudes can suffer greater ozone injury than those at lower altitudes due to the diurnal behavior of ozone. Though daily maximum and mid-day ozone concentrations are similar at different altitudes, the dosage increases with height. Ozone is rapidly depleted near the surface below the nocturnal inversion layer; however, mountainous sites above the nocturnal inversion layer do not experience this depletion. Therefore, the total exposure to ozone in mountainous areas can be much higher than in nearby valleys. Maximum ozone concentrations observed at elevated mountainous sites, as well as nonmountainous rural and remote sites often occur at night. In addition, sites at higher elevations are often exposed to sustained or multiple peak concentrations of ozone within a given 24-hour period. When assessing exposure-response relationships of forest ecosystems at high altitudes these considerations need to be taken into account. Most studies of forest ecosystems have monitored only the daylight ozone concentrations.

An additional factor that makes determining the effects of pollutant stress on ecosystems difficult, is that mature ecosystems are not completely stable, but maintain themselves in an oscillating steady state. Severe competitional among trees for light, water, and mineral nutrients results in the elimination of old and suppressed trees and the addition of new ones.

The elimination of the trees most susceptible to competitional stresses is accelerated by additional and severe periodic stresses such as drought, flooding, fire and attacks by insects or disease-causing organisms. Succession results, causing changes within an ecosystem. The effects of natural stresses upon ecosystems, unless they are catastrophic (e.g., fire, flood, or windstorm) are frequently difficult to determine.

Pollutant stresses, regardless of whether they are foliage or soil mediated, are superimposed upon naturally occurring stresses and may accelerate the processes of change already underway in ecosystems. Gaseous pollutant stresses, whether severe episodic stresses (one or more days of acute high exposure) or long-term chronic stresses, occur chiefly through the foliage.

Ozone is known to impair the growth of agricultural crops, and of native vegetation in ecosystems throughout the United States, more than any other air pollutant. Exposure of vegetation to ozone inhibits photosynthesis, alters carbon allocation and interferes with mycorrhizal formation in tree roots. Disruption of these important physiological processes can suppress the growth of trees, shrubs, and herbaceous vegetation by decreasing their capacity to form carbon compounds needed for growth and their ability to absorb the water and nutrients they require for life from the soil. In addition, loss of vigor increases susceptibility of trees to insect and pathogens and reduces their capability to reproduce.

It is not possible to state unequivocally what the effects of airborne pollutant chemicals on forest ecosystems have been. Stresses on trees from these pollutants have been both above ground through the foliage and through the soil. The responses of decreased growth, dieback and mortality observed at the present time in the forests of eastern North America are the result of long-term pollutant exposures and other multiple stresses upon individual trees and other perennial species. The effects of continuing pollutant stresses are cumulative and appear as growth responses. Growth responses take time. Such responses in perennial woody plants usually are not observable during the year they occur but often require a generation. This is particularly true of injury resulting from repeated acute episodic ozone exposures because there is time for recovery between peaks. It is possible, therefore, that the growth reductions in forest trees observable today in the eastern United States and attributed to many different factors are the result of physiological changes to which episodic ozone exposures that began many years ago were a major contributor.

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An evaluation of the role of ozone, acid deposition, and other airborne pollutants in the forests of eastern North America. Gen. Tech. Rep. SE-59. Asheville, NC: U.S. Department of Agriculture, Forest Service, Southeastern Forest Experiment Station; 1989. 172 pp.

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